ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

Winter 1963-1964

FEATURE ARTICLE:

Inhibition of Corrosion by the Pertechnetate Ion

● VOLUME 1

● NUMBER 2

TECHNICAL PROGRESS REVIEWS

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ISOTOPES AND RADIATION TECHNOLOGY

A REVIEW OF RECENT DEVELOPMENTS

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

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Foreword

Isotopes and Radiation Technology is a Technical Progress Review prepared by the Isotopes Development Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotopic power development, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore, readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

P. S. BAKER, ROBERT H. LAFFERTY, JR., and MARTHA GERRARD, Editors
A. F. RUPP, Advisory Editor
Oak Ridge National Laboratory

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by G. H. Cartledge, Chemistry Division, ORNL

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FEATURE ARTICLE

Inhibition of Corrosion by the Pertechnetate Ion

By G. H. Cartledge*

(Editor's Note: One of the most interesting properties of technetium is the "anticorrosive" characteristic of its pertechnetate ion,[1] TcO_4^-. Dr. Cartledge, a pioneer in this field, has written the accompanying article to bring up to date the status of the theories behind this particular use of technetium. The timeliness of such a report is emphasized by the fact that in the United States rusting annually consumes an amount of iron and steel equivalent to a quarter of the country's annual production.[2]

Technetium was not discovered until 1937, although many investigators, starting with Osann and his "polinium" in 1828, claimed to have isolated it.[3] However, in 1937, Perrier and Segrè irradiated molybdenum in a cyclotron and reported Tc^{95m} and Tc^{97m} with half-lives of 80 and 90 days, respectively.[4] In 1939, Seaborg and Segrè reported[5] 6-hr Tc^{99m}, which decayed to a long-lived isomeric form. This long-lived form, now known to be Tc^{99} with a half-life of 2×10^5 years, has been isolated from fission-product wastes. A few years ago it was priced at from \$2800 to \$1600 per gram, depending upon quantity, but can now be purchased from the Isotopes Sales Department of Oak Ridge National Laboratory for less than \$100 per gram).

Experiments have demonstrated that very effective inhibition of the corrosion of iron and steel can be achieved with low concentrations of pertechnetates under suitable conditions. Although there may be special circumstances in which the inhibitor may find practical application, its radioactivity and cost must necessarily restrict its widespread use. There is need, however, for further investigation of possibilities of using it in protective coatings or similar applications and also of its relation to the corrosion of nonferrous metals. The distinctive properties of technetium compounds make the element an excellent new research tool and differentiate it sharply from chromium, which forms the inorganic compounds so extensively used as inhibitors.

Properties of Technetium Compounds Related to Inhibition

A comprehensive review of the chemistry of technetium has been published recently,[6] but the following comparison with the chemistry of chromium will suffice to illustrate the differences that are significant for studies in inhibition.

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The important valence states of chromium and technetium are Cr(VI), Cr(III), Tc(VII), and Tc(IV). The bivalent chromate ion has basic and buffering properties,

$$\mathrm{HCrO_4^- \rightleftharpoons H^+ + CrO_4^{2-}}\ (K = 3.0 \times 10^{-7}) \qquad (1)$$

whereas the univalent pertechnetate ion is derived from a strong acid and lacks buffer capacity. When used at $10^{-3}\,f$ concentration and pH 6.0, the respective reversible electrode potentials are

$$\mathrm{CrO_4^{2-} + 5H^+ + 3}e^- \rightarrow \mathrm{Cr(OH)_{3(ppt)} + H_2O}; \quad E_h = 0.59 \text{ volt} \qquad (2)$$

and

$$\mathrm{TcO_4^- + 4H^+ + 3}e^- \rightarrow \mathrm{Tc(OH)_{4(ppt)}}; \quad E_h = 0.20 \text{ volt} \qquad (3)$$

Polarization measurements at a passive iron electrode indicate that the exchange current density of the pertechnetate electrode under the conditions specified above is several orders of magnitude greater than that of the chromate electrode. The pertechnetate ion, unlike the chromate ion, does not oxidize ferrous ions in weakly acidic solution, although it is effective in inhibition under the same conditions. Oxidation does occur, however, in alkaline solution.

The long half-life of $\mathrm{Tc^{99}}$ (2.15×10^5 years) and its 0.29-Mev beta radiation make it eminently suitable for tracer or autoradiographic techniques. At the low concentrations required in studies of inhibition, the radiation hazard is of no consequence. The strong absorption bands of the aqueous pertechnetate ion at 2440 and 2875 A are very useful for analytical purposes, the extinction coefficients being 6080 and 2310, respectively. The best value of the ratio of these coefficients is 2.63, and this is a useful test of the purity of pertechnetate solutions. The only electrode potential that has been directly measured[7] is that of the couple

$$\mathrm{TcO_4^- + 4H^+ + 3}e^- \rightarrow \mathrm{Tc(OH)_{4(ppt)}}; \quad E_h = 0.738 \text{ volt} \qquad (4)$$

Other important measured or calculated quantities are[8]

$$\text{Solubility of } \mathrm{KTcO_4} \text{ at } 25^\circ\mathrm{C} = 0.1057M$$

$$S^\circ \text{ for } \mathrm{TcO_4^-} = 47.9 \pm 0.3 \text{ eu}$$

$$\Delta S^\circ \text{ for } \mathrm{TcO_{4(aq)}^-} = -73.3 \text{ eu}$$

$$\Delta F^\circ \text{ for } \mathrm{Tc(OH)_{4(ppt)}} = -202.3 \text{ kcal/mole}$$

$$\Delta H^\circ \text{ for } \mathrm{Tc(OH)_{4(ppt)}} = -246.3 \text{ kcal/mole}$$

Technetium is recovered from fission products by solvent-extraction procedures and converted[9] to the salts $\mathrm{NH_4TcO_4}$ and $\mathrm{KTcO_4}$. Metallic technetium is obtained in the form of powder by a two-stage reduction of $\mathrm{NH_4TcO_4}$ by hydrogen. In the first stage $\mathrm{TcO_2}$ is produced at 200°C, and this is converted to metal at 700 to 800°C. The powder may be melted in an induction furnace (m.p. 2150°C). A few iron-technetium alloys have been prepared for metallurgical and electrochemical study.

Studies of Inhibition

When iron or low-carbon steel is immersed in a solution of potassium or ammonium pertechnetate at pH 5 or higher, there is an initial reaction wherein the inhibitor ion is reduced with formation of a film of mixed iron and technetium hydroxides. The extent of this reaction depends somewhat on the composition and surface activity of the metal; it may be measured by counting the beta activity of the water-rinsed specimen since absorption of the weak beta particles is slight in the thin film produced. The precipitated $\mathrm{Tc(OH)_4}$ can be reduced to much less than a monomolecular layer if an oxidizing agent, such as hydrogen peroxide, is added to the pertechnetate solution. Once passivation is achieved, no further consumption of the inhibitor is required if the solution is exposed to air. Two specimens of S.A.E. 1010 carbon steel have been immersed in a $5 \times 10^{-4}\,f$ solution since January 1953 (Fig. 1). The beta activity of the rinsed specimens has been measured 30 times over the 10-year period. It rose to a maximum within 2 hr after exposure at 95°C, and then slowly fell about 10% owing to reoxidation of $\mathrm{Tc(OH)_4}$ by oxygen. After the activity leveled off, the counts on one specimen showed the presence of 0.146 ± 0.005 μg of technetium on 4 $\mathrm{cm^2}$ throughout the long experiment. The surface of the specimens is still bright, the solution is clear, and the weight is unchanged within 0.1 mg.

The film of reaction products first formed is not sufficient for maintaining resistance to cor-

(*a*) (*b*)

Fig. 1 Uninhibited and inhibited steel corrosion. The specimens contain 10 ppm of chloride ion. (*a*) Corrosion in distilled water. (*b*) Inhibition of corrosion by pertechnetate ions.

rosion, however, even when heavy deposits are induced. Some minimum concentration of pertechnetate ions must remain in contact with the metal for inhibition to persist. This seems to be about 5 ppm of technetium ($5 \times 10^{-5} f$ TcO_4^-) with electrolytic iron and somewhat higher with carbon steels or cast iron.

Tests demonstrated that the perrhenate ion, ReO_4^-, which is similar in charge, size, and geometry, has no inhibiting properties at any concentration at temperatures between 5 and 95°C. A radioactive form of the perrhenate ion containing $Re^{186,188}$ was prepared to determine whether the radiation or the radiolytic products from technetium were involved in its action. When this was used at a specific activity corresponding to that of the inhibiting pertechnetate solutions, there was still no inhibition.

Experiments in which foreign ions, such as sulfate, were added to inhibited systems indicated that adsorption is involved in the overall corrosion-inhibition process. All the XO_4^{n-} inhibitors fail when the added ions exceed some concentration, which increases as the concentration of the inhibitor is increased.

The failure of the perrhenate ion to inhibit corrosion is particularly significant for adsorption theories since it is externally similar to the pertechnetate ion. Since the electrochemical polarization theory ascribes the inhibiting properties of chromate and, presumably, pertechnetate ions to their reducibility, polarization

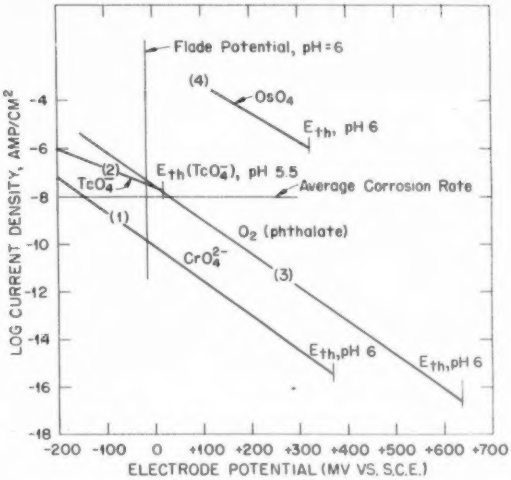

Fig. 2 Summary of polarization data at 25°C. Relative reduction rates of O_2, OsO_4, TcO_4^-, and CrO_4^{2-} on a passive-iron electrode at about *p*H 6 and 24°C: E_{th} represents the calculated reversible potential for each couple. [Reprinted by permission from *J. Phys. Chem.*, 65: 1367 (1961).]

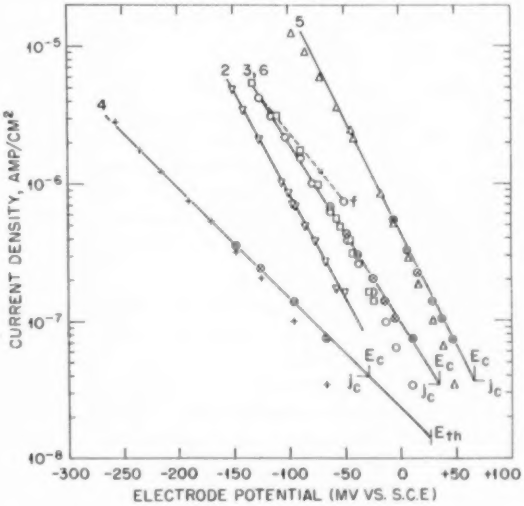

Fig. 3 Cathodic polarization of passive iron. Successive polarizations of passive iron in phthalate or pertechnetate solutions (0.010 *f* at *p*H 5.6 to 5.9). 2, oxygenated phthalate alone; 3, oxygenated pertechnetate, points ○, ending at *f*; 4, pertechnetate in helium, after point *f*; 5, oxygenated pertechnetate with $Tc(OH)_4$ left on film; 6, same after removal of $Tc(OH)_4$ by anodic polarization, points □; at lowest current densities, the data points require correction for an anodic density of 4×10^{-8} amp/cm², points ⊗. [Reprinted by permission from *J. Phys. Chem.*, 64: 1884 (1960).]

measurements have been made to determine the relative reduction rates of oxygen and reducible inhibitors on a passive iron electrode. Experimental results[10] for oxygen, technetium, and two other inhibitors (Fig. 2) show that the only inhibitor that exceeds oxygen in its rate of reduction at potentials in the neighborhood of the Flade (passivating) potential is OsO_4. These measurements disclosed an interesting feature of the electrodes exposed to pertechnetate ion and Os(VIII) oxide. In both cases the reduction products, $\mathrm{Tc(OH)}_4$ and $\mathrm{Os(OH)}_4$, accelerated the cathodic process, whether oxygen or inhibitor was being reduced. The data from an experiment[11] involving the reduction of pertechnetate ions (Fig. 3) again show the favorable properties of technetium compounds. Any $\mathrm{Tc(OH)}_4$ cathodically produced on the surface can be removed easily by brief anodic polarization so that effects associated with its presence can be identified.

Theories of Inhibition

In modern studies of the mechanism of corrosion and its inhibition, the radioisotope principally used has been Cr^{51}, with a 27.8-day half-life. This nuclide has been used, for example, in studies of the composition of passive films and the rate of deposition of such films on iron. Since the chromate ion is the most useful of the inorganic inhibitors of the passivating type, it is largely around this ion that theories of the action of such inhibitors have been developed.

These theories have considered rather varied conceptions as to the function of the inhibitor, the simplest being that the buffering and oxidizing properties of the chromate ion permit it to oxidize ferrous ions emerging through imperfections in the film and thus to precipitate a plug directly in the pore or crack. Evans,[12] who advanced this hypothesis years ago, now attempts to give a more generalized description of the conditions that give rise either to corrosion or to the formation of a protective film. For this purpose he considers the nature of the barrier to anodic dissolution which is interposed by whatever species is preferentially adsorbed on the metal surface from the solution. If this is water or hydroxide ions, emerging ferrous ions first meet the oxygen (negative) side of the adsorbed layer, and further migration is impeded by the more remote (positive) side of the layer. Protons are then ejected and carry the anodic current away from the surface, leaving an oxide film that may be protective. Evans then treats the chromate ion analogously, postulating that both it and the sulfate ion may have a positive region (XO_2^{2+}) analogous to that shown by the nitronium ion, as in crystalline $\mathrm{NO}_2^+\mathrm{ClO}_4^-$. Both Uhlig at MIT and Kolotyrkin in Russia have postulated electrical effects at the surface due to adsorption, but without developing the concepts to the degree that would permit a real differentiation between inhibiting and noninhibiting species.

Another theory[13] of the action of reducible inhibitors like the chromate ion is that reduction can occur at a sufficiently high current density and potential to polarize the metal anodically above that potential required for passivation. In this theory no specific properties are assigned to the inhibitor other than its oxidation-reduction potential and exchange current density. Although this explanation may suffice in particular cases, it takes no account of specific interactions between the surface and aqueous ions or molecules that are present. Furthermore, the theory does not explain how certain nonoxidizing inhibitors make it possible for oxygen to induce passivity in the presence of the inhibitors, whereas oxygen would accelerate corrosion in the absence of the inhibitors. The polarization theory, without supplementary considerations, cannot therefore be considered a general theory of inhibition.

There are also reasons why oxidation and buffering do not appear to be sufficient as a general explanation of inhibition.[14] Further, the ideas regarding surface effects of adsorption were unable to explain why chromate ions inhibit corrosion and sulfate ions do not, although the two ions have the same charge, geometrical configuration, and nearly the same size. It was this situation that led to the suggestion that the differentiation must lie *within* the ion, rather than in its gross charge, size, or geometry.

A crude electrostatic calculation[14] made in 1952 showed that chromate and sulfate ions will induce space charges of opposite polarity when adsorbed on a dielectric surface if, as seemed likely, the sulfur-oxygen bonds are essentially covalent and the chromium-oxygen bonds are largely ionic. This difference in polarity should affect the kinetics of charge transfer across the interface. If the space charge is indeed related to the difference in corrosion rates, it should then be possible to describe conditions favorable to inhibitory properties in the XO_4^{n-} structure: (1) a high central positive charge, (2) ionic bonding, X-O, and (3) an oxidation-reduction potential low enough to prevent rapid reduction of the XO_4^{n-} particles past the point of forming a surface film of reaction products. The last criterion excluded the powerful oxidant MnO_4^-, and it was therefore apparent[15] that technetium, in the seventh group of the periodic system, was the most likely of all elements to have the desired combination of properties. There were reasons for anticipating that the ReO_4^- ion would be less inhibitive, but no way to predict how much less.

Experiments[16] with electrolytic iron and carbon steel in November 1952 immediately demonstrated the correctness of the prediction regarding the pertectnetate ion. It was found to be more effective both at lower concentrations and at higher temperatures than the chromate ion. Thus $5 \times 10^{-5}\,f$ $KTcO_4$ gave excellent protection to S.A.E. 1010 steel at 250°C in a test lasting 116 hr; a visible film formed on the specimen, but the solution remained clear, and the weight of the metal was unchanged within 0.1 mg.

Summary

The experiments cited illustrate how technetium may be used to evaluate the various theories of inhibitor action. For example, the absence of buffer action and the mild oxidation potential argue against the early chemical hypothesis. The extreme contrast between the characteristics of TcO_4^- and ReO_4^- disproves any idea of adsorption that considers only the external features of the ions. The theory of purely electrochemical polarization is doubtless a valid description of action that could account for passivation, although the measurement of the comparative contributions of oxygen and reducible inhibitors shows that oxygen is the chief source of cathodic current with either TcO_4^- or CrO_4^{2-} as an inhibitor. The action of sulfate ions in destroying inhibition also shows the incompleteness of the polarization hypothesis.

There is considerable evidence to support the assumption of the electrostatic polarization hypothesis, which looks inside the various ions for differentiating characteristics among them.[14,17] The conclusion most likely to be reached ultimately is that each of the ideas thus far proposed is meaningful for some aspect of the inhibitory process, the complete understanding of which will necessitate the combination of the points of view of the electrochemist and the solid-state physicist.

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Section I

Isotopes and Radiation Technology

Isotope Production and Development

Radioisotopes may be produced in several ways: (1) by activation of stable nuclides in a reactor or in an accelerator such as a cyclotron; (2) by induced fission, the fission products being produced either deliberately or as by-products; and (3) by spontaneous fission. This section describes the Oak Ridge National Laboratory (ORNL) facilities for both reactor and cyclotron production of radioisotopes and reviews briefly the production of the important isotope I^{125} by both methods. Also presented here is a table of the stable fission-product isotopes which, because of their unusual isotopic distribution, can be used advantageously as starting materials to provide enriched target materials for radioisotope production. Some of the advantages of using enriched stable isotopes as targets for radioisotope production in cyclotrons and reactors were covered in an earlier report.[1] Finally, the current status of "milker" systems is reviewed, in which parent nuclides with convenient half-lives decay under such conditions that the useful daughter radioisotopes are obtained by simple chemical processing.

The number of nuclides available from the numerous reactors in the United States[2] and throughout the world has grown until more than 100 can be considered stock items, and many hundreds more can be made to order. From small natural-uranium graphite reactors to high-flux research reactors and power reactors using enriched uranium or plutonium, the number of different radionuclides that can be produced by (n,γ), (n,p), (n,α), and $(n,$ fission$)$ reactions is largely a matter of space, flux, and demand. For example, ORNL alone provides about 90 reactor-activated isotopes, 53 of which are (n,γ), 22 $(n,$ fission$)$, 6 (n,p), and 2 (n,α) reactions.[3] The Institutt for Atomenergi at Kjeller, Norway, offers about 30 isotopes, most of which are produced in their reactor JEEP.[4] The Radiochemical Centre at Amersham, England, provides an extensive range of isotopes and labeled compounds prepared in their reactors.[5] Similarly, Atomic Energy of Canada Limited;[6] Le Commissariat à l'Énergie Atomique (France), Le Centre d'Étude de l'Énergie Nucléaire (Belgium),[7] the Soviet Union,[8] and others provide isotopes and services.[9]

Many more curies of isotopes are produced in reactors than in accelerators. Except for fission products, Co^{60} alone, which is reactor produced by neutron irradiation of Co^{59}, accounts for more activity than all other isotopes combined. Historically, however, radioisotope production in accelerators, such as cyclotrons, predates reactor-activated isotopes by many years. For example, tracer amounts of P^{32} were produced[10] in cyclotrons as early as 1935, whereas reactor-produced P^{32} and C^{14} did not become available until 1946, when the general AEC program of producing isotopes in reactors was initiated at ORNL.[11] Among the other early accelerator-produced isotopes were C^{14}, Ca^{45}, K^{42}, Br^{80}, Cl^{38}, and Na^{24} (Ref. 12). Accelerators currently play an extremely important production role because of the versatility of the machines, the variety of reactions possible, and the fact that most isotope products are neutron deficient, a state not easily achieved in reactors. Furthermore, when desired, neutrons or other particles of various energies can be produced with accelerators, making them highly suitable for activation analysis[13] as well as for isotope production.

Oak Ridge Research Reactor for Isotope Production

By J. J. Pinajian

The Oak Ridge Research Reactor (ORR),[14] completed in March 1958, and its associated facilities are described here. With the permanent shutdown of the 20-year-old graphite reactor on Nov. 4, 1963, the ORNL facilities for reactor production of isotopes are the ORR, with a maximum thermal flux of $\sim 5 \times 10^{14}$ neutrons/(cm^2)(sec), and the Low-Intensity Test Reactor (LITR), with a flux of $\sim 2 \times 10^{13}$ neutrons/(cm^2)(sec). The ORR is generally more suitable for radioisotope production since it permits higher specific activities, or smaller amounts of target, or both.

The ORR is a water-cooled and -moderated Materials Testing Reactor (MTR) type research reactor, which uses enriched uranium in the form of boxed elements filled with curved fuel plates and beryllium reflector pieces in a seven- by nine-element rectangular lattice.[15] The boxed elements of fuel and beryllium comprise the reactor core, which is 21 by 27 by 24 in. The fuel elements are parallel plates of aluminum sandwiching a uranium-aluminum alloy, bowed to increase their strength against buckling.[16] The 19 parallel plates are packed closely so that, from the nuclear standpoint, the system is essentially homogeneous.[17] The reactor characteristics[18] of interest in the radioisotope program are as follows:

Operating power	30 Mw
Thermal-neutron flux	1.6×10^{14} neutrons/(cm^2)(sec), average; 5×10^{14} neutrons/(cm^2)(sec), maximum

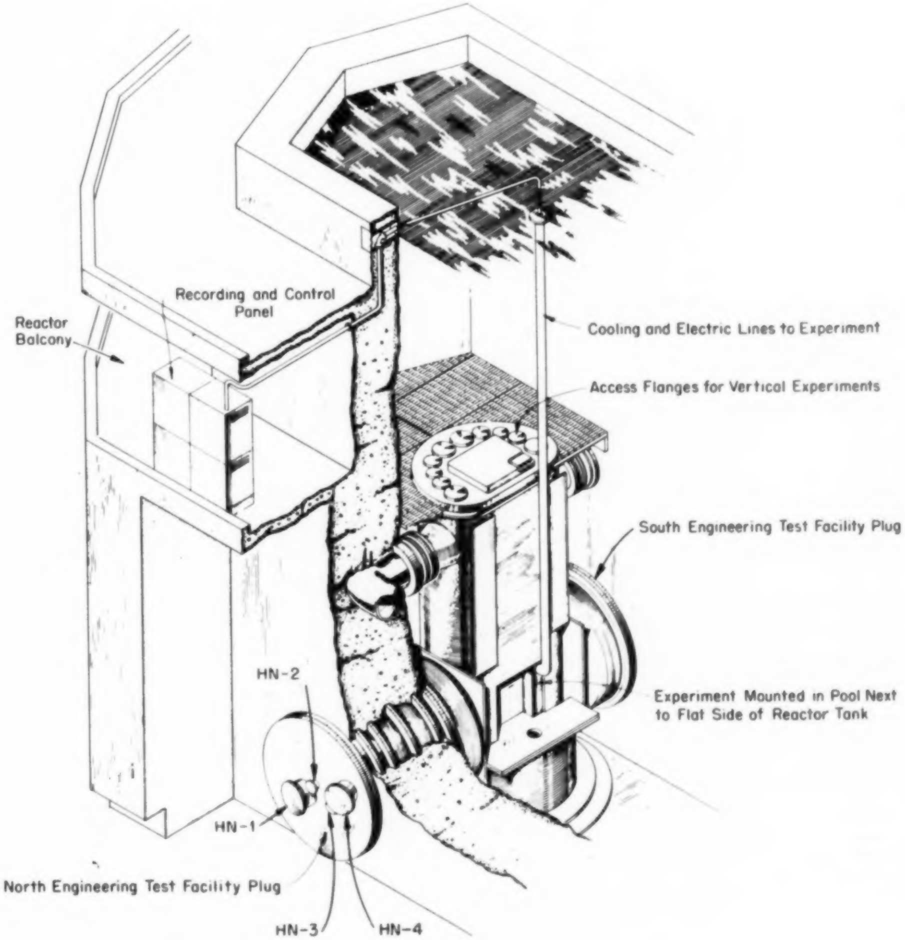

Fig. I-1 ORR engineering test and poolside irradiation facilities.

Fast-neutron flux	Varies according to position in lattice
Operating cycle	8 weeks, comprising 7 weeks at full power and 1 week of shutdown for maintenance, experimental changes, and refueling
Refueling cycle	~12 days; there are three half-day shutdowns for refueling within each operating cycle
Operating time	~80%
Unscheduled shutdowns	About four per month (average)
Gamma heating	3 to 10 watts/g, depending on material and lattice position

The reactor core tank, the poolside irradiation facilities, and the engineering test facilities are shown in Fig. I-1. Figure I-2 is a drawing of the core and tank.

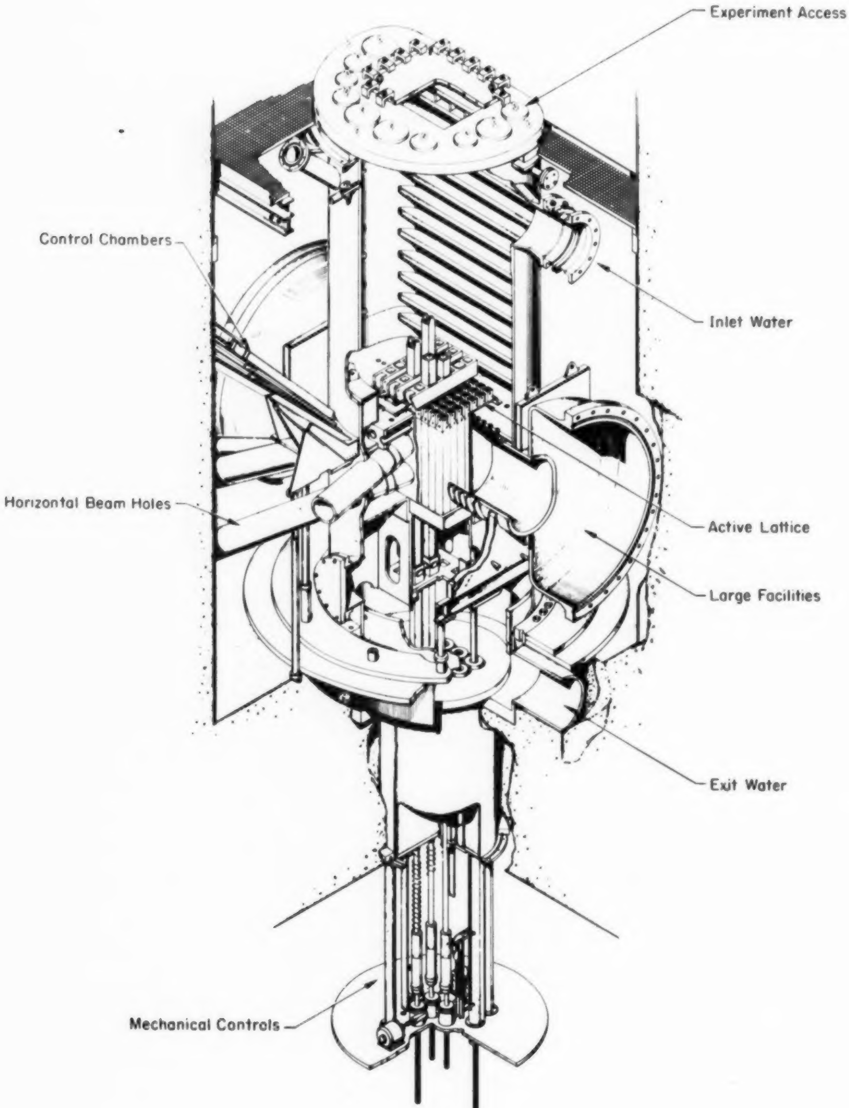

Fig. I-2 View of the ORR core and tank.

Irradiation Facilities

The irradiation facilities available for routine use by the Isotopes Development Center are shown in a typical lattice pattern in Fig. I-3. Access is through tubes (V-1, V-2, . . . , V-11) in flanges in the reactor tank top (Figs. I-1 and I-2). The lattice positions are identified by a combination of a letter (A through G) and a numeral (1 through 9). For example, in the lattice pattern shown, hole V-4, which gives access to lattice position F-8, is a hydraulic tube; a second hydraulic tube, V-3, feeds position E-9. The hydraulic system with its loading and unloading system, a lattice element containing the hydraulic tube, and the hydraulic rabbit are shown in Fig. I-4. Seven core lattice positions with a flux of $\leq 5 \times 10^{14}$ neutrons/(cm^2)(sec) are used by the Isotopes Development Center:

1 to 3. Removal stringers in positions C-2, F-5, and D-9. Two of these will accommodate 80 samples each and the other, 60 samples. Figure I-5 shows an 80-hole stringer partially loaded with the aluminum irradiation cans, which are 0.375 in. in diameter and 2.562 in. long and have a 0.032-in.-thick wall. These cans are sealed by welding and are leak tested before insertion in the reactor. The trays are noted by letters Q (starting at the top) through Z, and the positions are noted by numerals 1 (at the right of the pivot) clockwise through 7, with number 8 at the center. Materials are irradiated in three-week cycles or multiples thereof. The stringer is loaded and unloaded with tongs to take advantage of the water as shielding material.

4. The NF (fast stringêr) at position A-3. A short middle section containing nine sample holes and two large end sections containing three large holes connect to form a fuel-element-shaped unit. The facility is used with a beryllium reflector and has the best available fast flux for (n,p) reactions [e.g., $S^{32}(n,p)P^{32}$]. It is also used when a large volume is required [e.g., $Xe^{124}(n,\gamma)Xe^{125} \xrightarrow{\beta} I^{125}$].

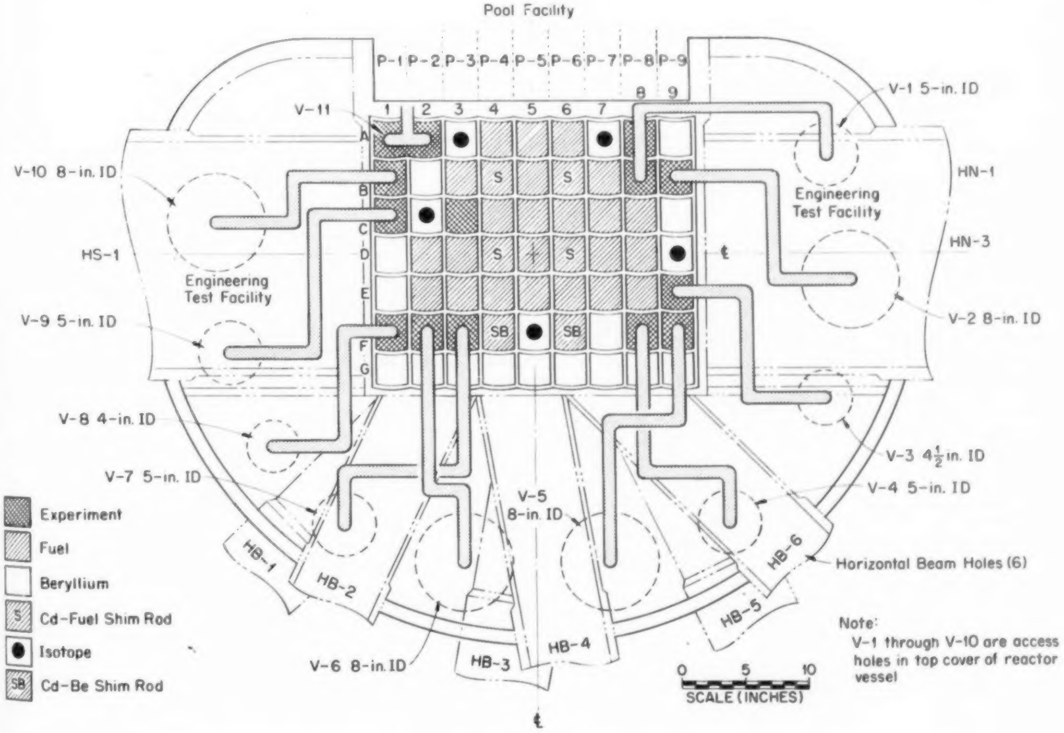

Fig. I-3 ORR lattice pattern and experiment locations.

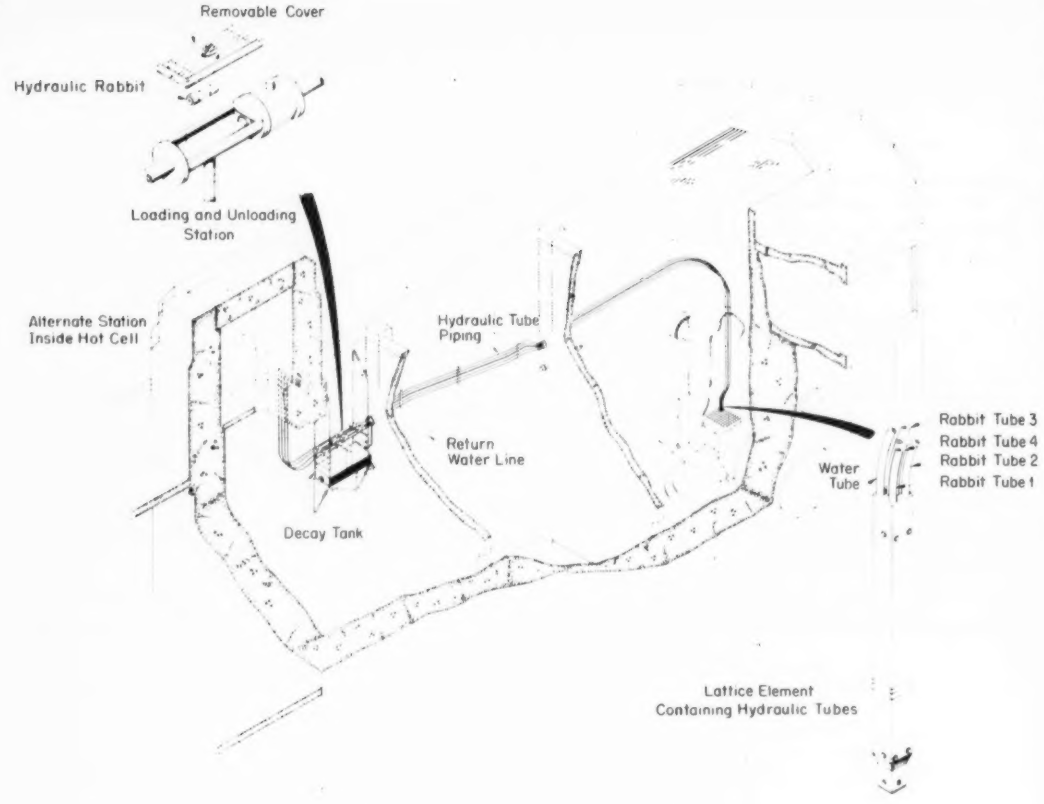

Fig. I-4 ORR hydraulic rabbit system.

Fig. I-5 Partially loaded 80-hole stringer.

Table I-1 PROPERTIES OF SOME FAST-FLUX MONITORS

Material	Reaction	Half-life	Effective threshold,* Mev
P^{31}	$(n,p)Si^{31}$	2.6 hr	2.5
S^{32}	$(n,p)P^{32}$	14.3 days	2.9
Ni^{58}	$(n,p)Co^{58}$	72 days	5.0
Al^{27}	$(n,p)Mg^{27}$	9.8 min	5.3
Si^{28}	$(n,p)Al^{28}$	2.3 min	6.1
Mg^{24}	$(n,p)Na^{24}$	15.0 hr	6.3
Al^{27}	$(n,\alpha)Na^{24}$	15.0 hr	8.6

*From Ref. 20.

5. The I^{131} assembly at position A-7. It consists of a series of aluminum-clad uranium-aluminum alloy rings arranged in a fuel-element-shaped unit. Iodine-131 is produced as a fission product.

6 and 7. The hydraulic tube facility, in lattice positions F-8 and E-9, accessible through vents V-4 and V-3, respectively. Three of the four tubes at position F-8, each of which is available for irradiations of ≤7 days, are now being used by the Isotopes Development Center. Up to three samples, or "rabbits," may be loaded in each tube. The thermal-neutron flux in lattice position F-8 is 1 to 2×10^{14} neutrons/(cm^2)(sec). The irradiated rabbits can be unloaded alternatively inside a hot cell, if desired.

In addition, the pool irradiation facility (outside the tank) has available a maximum thermal and fast (>0.4 Mev) neutron flux of $\sim 4 \times 10^{13}$ neutrons/(cm^2)(sec).

Neutron-Flux Measurements

The experiments being conducted in the ORR are various and complex. Because of the small size of the core (21 by 27 by 24 in.), changes in experimental conditions and loading will result in changes in reactivity, neutron spectral distortions, and neutron flux. Materials with excessively high neutron capture or scattering cross sections will cause flux perturbations (i.e., changes in the neutron flux in adjacent areas) as well as flux depression (i.e., decrease of the neutron flux as a function of penetration

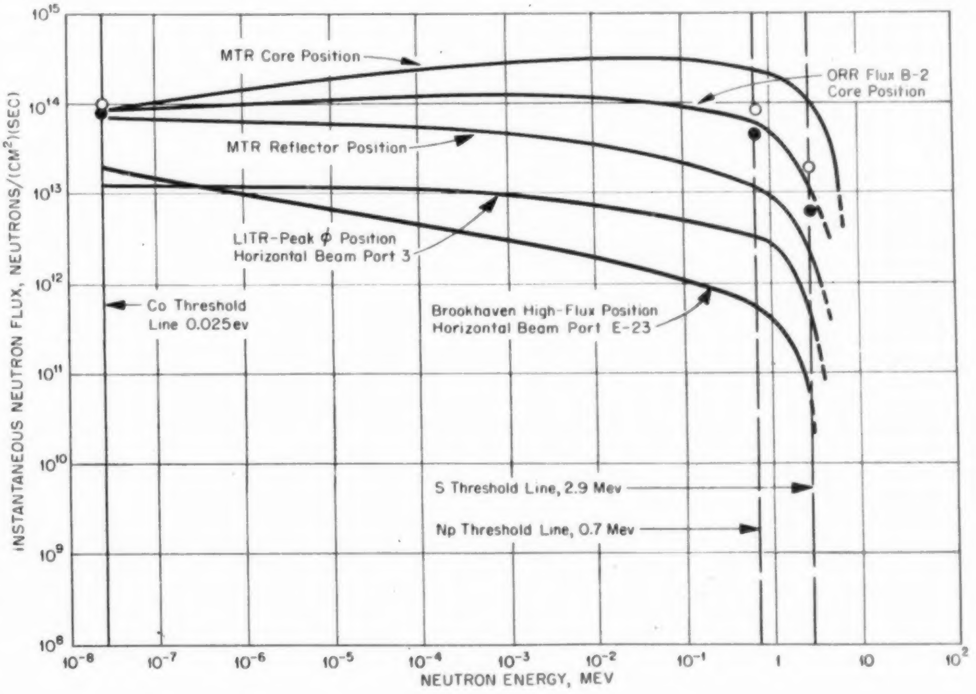

Fig. I-6 B-2 position spectral distribution shown compared with data obtained by Steele and Hawthorne.[20a] ○, maximum in B-2 position. ●, minimum in B-2 position.

into the target material itself and hence a nonuniform exposure of the target mass to the neutrons). Therefore flux measurements are unique values for each reactor loading arrangement and must be considered, within limits, as nonreproducible. To further complicate matters, there are two general conventions used in measuring and reporting neutron fluxes and threshold energies.[19] However, core loading profiles are kept, from which neutron-flux values for various "isotope" positions can be predicted with reasonable accuracy.

Table I-2 NEUTRON FLUX IN STRINGERS 1, 2, AND 3

(From 1554 Hr on Aug. 14, 1960, to 2100 Hr on Aug. 28, 1960. Monitor: Cobalt-Aluminum Alloy and Cobalt Wire; Power: 30 Mw)

Stringer 1 Core position C-2		Stringer 2 Core position F-5		Stringer 3 Core position D-9	
Tray and position	Flux, n/(cm^2)(sec)	Tray and position	Flux, n/(cm^2)(sec)	Tray and position	Flux, n/(cm^2)(sec)
Q-8	0.443×10^{14}	Q-6	0.662×10^{14}	Q-5	0.476×10^{14}
R-6	0.837	R-8	1.57	R-1	1.15
S-8	1.67	R-3	1.62	R-5	0.976
T-5	2.20	S-8	2.58	S-5	1.65
U-2	3.02	V-2	5.21	T-1	2.46
V-6	3.20	W-1	4.62	W-5	2.49
W-4	2.93	Y-8	3.49	X-1	2.57
X-8	2.89	Z-6	1.93	Y-5	1.74
Y-6	2.27	Z-8	1.99	Y-1	2.05
Z-8	1.62			Z-1	1.22

Although there are several ways of measuring the neutron flux, foil or wire activation is most convenient because of the limited space in the irradiation can.[19] Neutron-flux determination by the activation method relies on a knowledge of the reaction cross section of the monitor and subsequent analysis or measurement of the induced activity. The method can be used to measure thermal, resonance (or epithermal), and fast-neutron fluxes. Although characteristics of fast-neutron activation monitors are well known,[20] the average cross section varies from facility to facility because of differing neutron spectra. The effective threshold energy varies also.

Neutron-Flux Monitors. Thermal-neutron fluxes are generally measured with fine cobalt wire or with a cobalt-aluminum alloy (0.151 at.% cobalt). The alloy effectively disperses the cobalt throughout the aluminum so that the flux is not depressed within the cobalt. The use of cobalt wire requires a correction factor for the flux depression effect. A cross section of 36.7 barns is used for the $Co^{59}(n,\gamma)Co^{60}$ reaction.

Table I-1 lists several fast-neutron monitors which are suitable for use in the ORR and which can be readily fabricated into desired forms. The radiation characteristics, competing reactions, half-lives of the products, magnitude of the flux to be measured, and the temperature of the medium in which the monitor is to be placed dictate the choice of material.

Figure I-6 compares the spectral distribution in the B-2 core position measured by Dragoumis[21] (open circles represent a maximum, and closed circles, a minimum), with data obtained by Steele and Hawthorne.[20a] Table I-2 gives a typical flux picture of stringers 1, 2, and 3.

Summary

The ORR has a maximum flux of 5×10^{14} neutrons/(cm^2)(sec). Its facilities for irradiation include three removable stringers, which will accommodate a total of 220 samples; a fast-flux stringer, which may be used when large volumes are to be irradiated; the I^{131} assembly; a hydraulic tube facility; and a pool irradiation facility, which has a maximum thermal and fast (>0.4 Mev) neutron flux of $\sim 4 \times 10^{13}$ neutrons/(cm^2)(sec).

ORNL 86-in.-Cyclotron Facility for Isotope Production

By J. J. Pinajian and T. A. Butler

The ORNL 86-in. cyclotron, which is described here, has an important function of producing neutron-deficient radioisotopes. These radioisotopes usually give rise to (1) positron emission, which results in 0.511-Mev annihilation quanta, or (2) electron capture, which results in characteristic X-ray emission (one or the other of which is usually associated with decay of neutron-deficient radionuclides) as additional desirable features. In contrast to the convenient half-lives of the cyclotron-produced radioisotopes, the reactor-produced neutron-excess radioisotopes often have limited use because of their short half-lives.

As a producer of radioisotopes, the cyclotron complements the reactor by producing materials with extremely high specific activities. Also, certain elements have cyclotron-produced radioisotopes with more convenient half-lives or radiation properties than the radioisotopes produced in reactors. For example, Fe^{55}, Co^{57}, and Cd^{109} may be made with specific activities as high as 760, 7200, and 2000 curies/g, respectively. At the other extreme, a 0.001-μa 23-Mev proton beam from the cyclotron is used to simulate Van Allen belt radiation in National Aeronautics and Space Administration studies on the effect of radiation on space vehicle and satellite components. This irradiation service is also used by space biologists to determine radiation doses that may be survived by various organisms.

Construction was started on a fixed-frequency high-beam-intensity (1 ma) 19-Mev cyclotron[22] in September 1949. The first beam in this unusual accelerator was obtained in November 1950, and for two years it was used primarily for the production of Po^{208}. In early 1952, internal revisions in positioning and mounting the ion source resulted in an increase in proton energy from 19 to ~23 Mev. But, because of the 400- to 450-kv voltage between the dees (D-shaped hollow electrodes that supply the accelerating potential), additional cooling was required to compensate for radiofrequency heating in the upper portions of the machine. At the higher energy the Po^{208} yield was more than doubled. A total of ~9 curies of Po^{208} was

Table I-3 EXPERIMENTAL YIELDS OF CARRIER-FREE RADIOISOTOPES ROUTINELY PRODUCED IN THE CYCLOTRON

Radio-isotope	Half-life*	Target material	Yield, mc/hr			
			Flat plate	Capsule	Window	T-position
Be^7	53 days	Li on Cu	30			
		LiH		16†		
Na^{22}	2.58 years	Mg	0.35	0.04		
V^{48}	16.1 days	TiO_2		25		
Cr^{51}	27.8 days	V	120‡			
Mn^{52}	5.7 days	Cr		65		
Fe^{55}	2.6 days	Mn on Al	8			
Co^{57}	267 days	Ni on Cu	24		1	
Ge^{68}	280 days	Ga on Cu	3			
As^{74}	18 days	Ge		22		
Rb^{84}	33 days	Kr				1.1
Sr^{85}	64 days	RbCl		5		
Y^{88}	105 days	$SrCO_3$		5		
Tc^{95m}	60 days	Mo		2.5		
Cd^{109}	1.3 years	Ag	9			
Ce^{139}	140 days	La_2O_3	1.2			
W^{181}	145 days	Ta	13		0.5	
Au^{195}	180 days	Pt on Cu	8.5			
Bi^{207}	28 years	Pb on Cu	0.3			

*Half-lives are taken from Ref. 24.

†Oversize capsule.

‡Modified flat plate.

produced before the project was terminated[23] in August 1952.

During the next few years, the groundwork for the production of neutron-deficient radionuclides was laid, and the high-current flat-plate target, the intermediate-current capsule target, and the low-current window target were designed. A deflection system was installed in 1956 to deflect the proton beam through a pipe to targets 40 ft away.

Until recently the primary function of the machine was for investigations in nuclear physics research, and only limited production time was made available. Since early 1962 a large percentage of the time has been devoted to production. Experimental yields of carrier-free radioisotopes routinely produced in the cyclotron are shown in Table I-3.

The ORNL 86-in. fixed-frequency cyclotron (Fig. I-7) is a "vertical" cyclotron, the dees being suspended from above.[25] The installation (Fig. I-8) is provided with 5-ft-thick concrete shielding walls. A 400-ton magnet energized by a 1500-amp current supplies the 8800-gauss field. The deflection system is a magnetic channel coil energized with a 4300-amp current, which cancels the existing magnetic field and allows a partially deflected beam to continue out of the tank (Fig. I-9). Primarily because of a phase slip, the ions lag behind one another during the voltage change of the dees, and the outer orbits come closer and closer. The beam is extracted after ~25 revolutions, at which point the energy is 23 Mev, and the relativistic mass of the particle has increased ~2.6%.

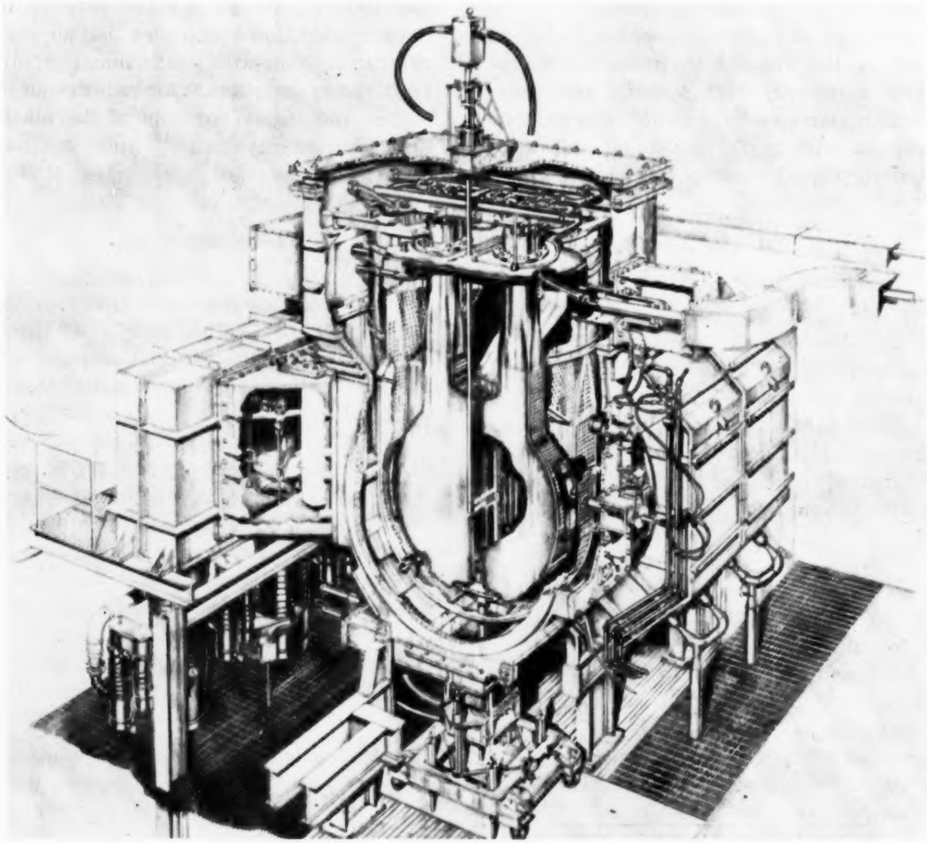

Fig. I-7 The ORNL 86-in. cyclotron.

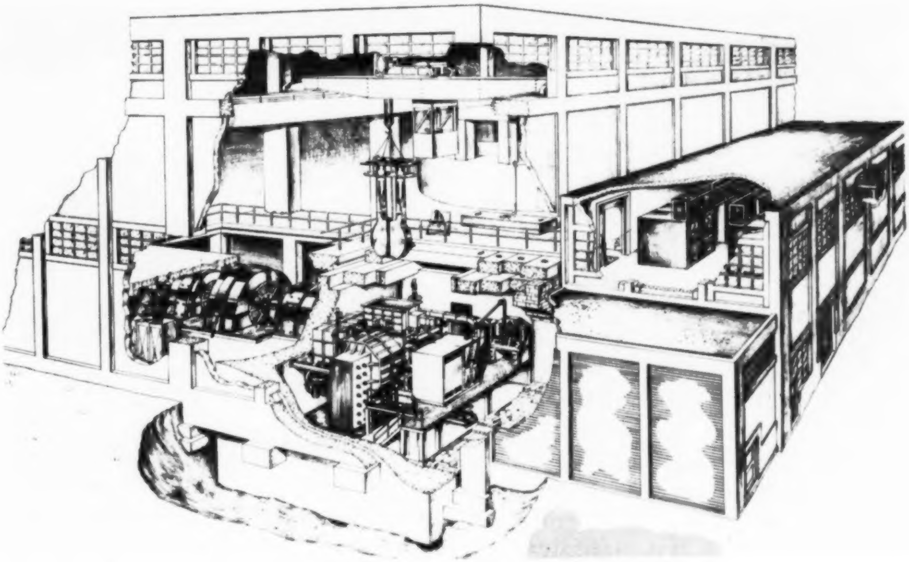

Fig. I-8 The ORNL 86-in.-cyclotron facility.

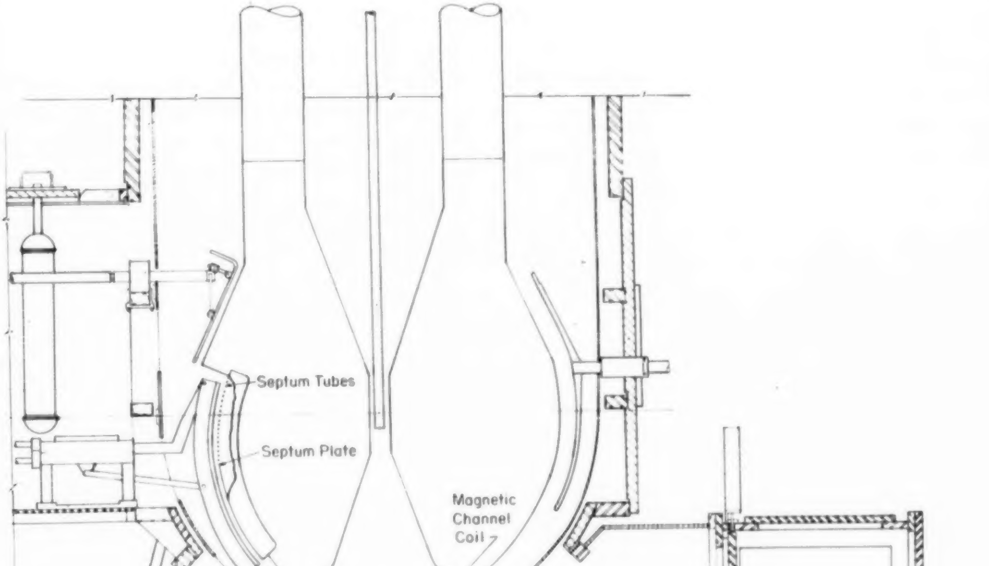

Fig. I-9 The 86-in.-beam deflection system.

Although the machine has a fixed frequency, there is a certain latitude of energy because of the possibility of varying the distance of the target from the center of the orbit. A proton energy range of 17 to 23 Mev is available, and the cyclotron is capable of developing and utilizing a 2600-μa proton current at the lower energy. Targets are loaded and unloaded through a vacuum lock at the bottom of the machine.

Irradiation Facilities and Targets

The configuration of the targets used in the 86-in. cyclotron is determined by the type of material used. Irradiation may be either internal or external.

Internal Beam, Flat-Plate Targets. The maximum internal beam current, ~2600 μa, requires the dissipation of ~60 kw from an area ~1.5 by 3.0 in. Such heat dissipation is possible only with suitable conductors such as solid copper or silver targets. Hence, for high beam currents, a pure-copper, flat-plate target (Fig. I-10) is used; it is $5\frac{5}{8}$ by 6 in. and 0.5 in. thick and has 15 to 23 cooling holes through which 50 to 80 gal/min of cooling water at 140 psi is passed. Metals that are easily electrodeposited, rolled, brazed, or cast are used with a copper or an aluminum base.

The amount of the beam current that is usable depends on the surface temperature, which must be kept below the melting or vaporizing point of the target. These currents vary from 225 μa for lithium metal cast in a recess of a copper flat-plate target to 2600 μa for solid-silver targets. Several curies of Fe^{55} and Co^{57} have been produced with flat-plate targets.

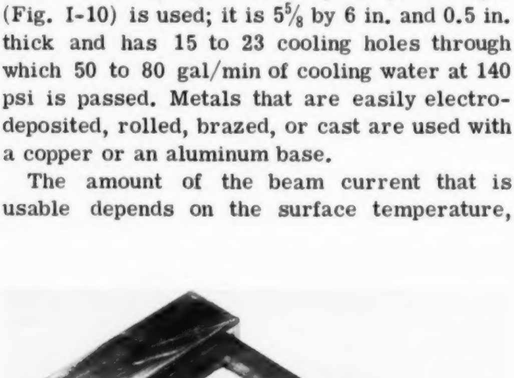

Fig. I-10 High-current flat-plate target partially assembled in header unit.

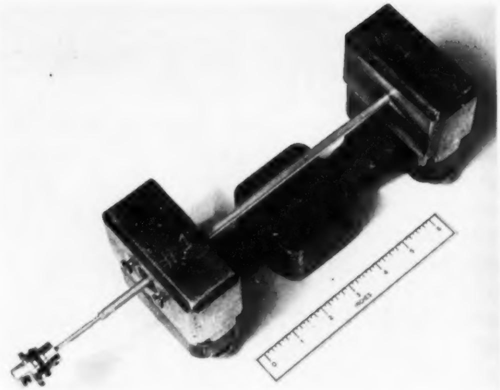

Fig. I-11 Capsule target partially assembled.

Internal Beam, Capsule Targets. Capsule targets[26] are used for irradiation of metal powders or chemical compounds, despite the fact that nearly one-third of the 190- to 200-μa beam is lost to the leading edge of the capsule and jacket. Stable oxides or other high-melting compounds are preferred as target materials since internal temperatures reached are greater than 1000°C. This type of target is economical for irradiation of scarce or expensive materials, such as enriched isotopes, rare earths, precious metals, and for irradiation of materials that yield gaseous products. The capsule, shown partially installed in its header unit in Fig. I-11, is an aluminum or a nickel tube with 0.010-in.-thick walls, 0.75-in. plugs welded in place at the ends, and a 3-in. void for the powdered target. The capsule is centered in the header so that water at 140 psi can flow through the 0.015-in. annular space. Materials routinely irradiated in capsule targets include TiO_2, chromium powder, RbCl, $SrCO_3$, La_2O_3, rare-earth oxides, and isotopically enriched substances. Several curies of As^{74} have been produced from germanium powder in this type of target.

Internal Beam, Window Targets. Isotopically enriched metal powder and chemical compounds

(5 to 50 mg) as well as foil strips can be irradiated in a target with a 0.010- to 0.012-in. window (Fig. I-12). High-energy and low-energy side reactions are practically eliminated by using absorbers and by decreasing the thickness of the target, respectively. The target materials are either wrapped in metallic foils (usually aluminum) or sealed in metallic tubes of aluminum, nickel, or platinum. A beam of 80 to 90 μa is normally used with water at 40 psi to cool the back of the target. Aluminum absorbers can be used to reduce the proton energy from the maximum of 23 Mev [for $(p,2n)$ and $(p,3n)$ reactions] to ~15 Mev [for (p,α) reactions] or to ~12 Mev [for (p,n) reactions]. Hence there is a certain degree of energy selectivity with this target.

Most irradiations of enriched material are carried out in targets of this type. The target is also economical when only small amounts of activity are needed.

Deflected Beam, T-Position Targets. About 12 ft from the cyclotron tank is a versatile facility to accommodate the <20-μa beam (Fig. I-13). In general, experimental amounts of short-lived radionuclides for research purposes can be conveniently and rapidly produced at this position.

Deflected Beam, 15°- and 0°-Position Targets. The deflected beam is passed through a pipe in the 5-ft-thick concrete wall and into a magnetic deflection system (Fig. I-14). A 15° deflection occurs when the magnet is operating, and the beam ($\ll 0.1$ μa) terminates in a scattering chamber. When the magnet is not energized, the protons continue along the straight pipe, and a 0.14-μa beam terminates 40 ft from the cyclotron tank. Radiation-damage studies and service irradiations of foil stacks are routinely performed. Nuclear reaction cross sections and thick-target yields are also measured at this position.

Research Program

A continuing program is being conducted to develop targets, production techniques, and chemical-processing procedures. In many instances, considerable effort is being devoted to obtaining high-purity targets and target backings or capsules. The preparation of Co^{57} with a specific activity greater than 6 curies/mg is an example of the successful completion of such work. High-specific-activity Co^{57} was made for workers labeling vitamin B_{12} and for investigators of the Mössbauer effect from nickel targets containing <0.24 ppm of nickel. The targets were made by electroplating nickel (purified by ion exchange) from solutions of nickel salts onto an electrode of nickel that was low in cobalt. Chemical problems in recovering the Co^{57} were solved by electrostripping[27] the cobalt and nickel from the face of the target, removing the copper by H_2S, and isolating the Co^{57} by ion exchange. Cobalt solutions are now routinely obtained with specific activities of >6 curies of Co^{57} per milligram of cobalt (>70% of theoretical), with evidence that the inactive cobalt atoms present may actually be produced in the cyclotron by nuclear reactions.

Fig. I-12 Window target. The filled and sealed tube in the foreground is held in the slotted window area by the bronze clamps.

High-specific-activity Fe^{55} has also been produced by perfecting manganese electroplating techniques specifically designed to eliminate iron impurities. Rubidium-84 for medical applications was produced from krypton gas depleted in Kr^{86}, which decreased production of of the unwanted Rb^{86}. This krypton, after irradiation, was collected for reuse in a Monel thimble immersed in a liquid-nitrogen trap, and the radioactive rubidium was washed out of the target chamber with hot distilled water.

At present the production of Pd^{103} from rhodium-plated targets is being investigated. An a-c electrodissolution method, developed for use with a number of relatively noble

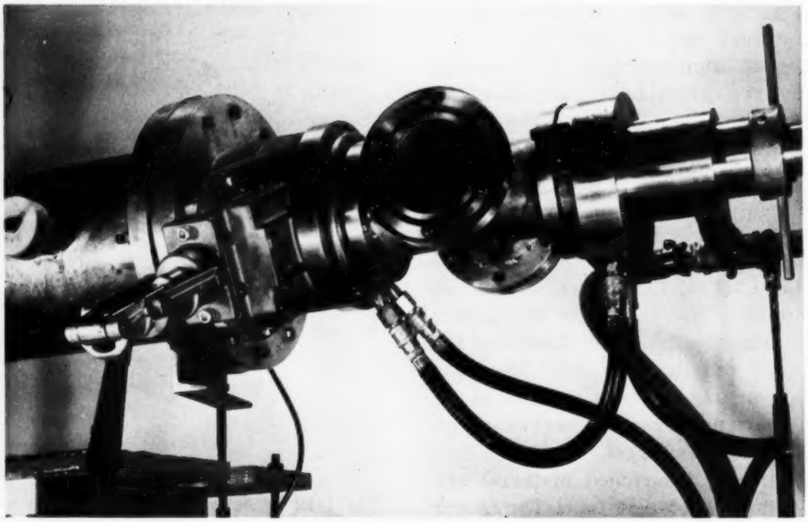

Fig. I-13 T-position target. The deflected beam enters on the left. The collimator cooling hoses are in front of and behind the Lucite-covered ports.

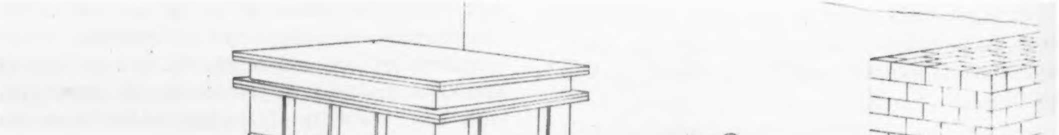

Fig. I-14 Deflector system installed in the 86-in. cyclotron.

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metals (e.g., platinum, rhodium, iridium, and tantalum), will be used to remove the irradiated rhodium and palladium from the targets.

Summary

The ORNL 86-in. cyclotron, which is used in producing radioactive isotopes with high specific activities, e.g., Co^{57}, Fe^{55}, and Pd^{103}, has a maximum proton-beam current of 2600 μa. The proton energy range is from 17 to 23 Mev. Irradiations may be made either internally or externally with various types of targets.

Iodine-125 Production

Iodine-125, whose preparation and fabrication into radiographic sources are reviewed here, is an isotope of considerable current interest. It was discovered in 1946 by Reid and Keston,[28] although mistaken by them for I^{129}; in 1947 it was indicated to be "more probably I^{125} than I^{129}" by Glendenin and Edwards[28a]; and it was ultimately confirmed as I^{125} by Friedlander and Orr.[29] The cost of this early cyclotron product, $2000 to $3500 per millicurie at that time, prohibited any practical use for it. However, efforts to develop a low-cost large-scale process were undertaken at the request of physicians, and cyclotron production[30] and processing[30a] costs were cut to a few hundred dollars per millicurie. In 1951 Bergström[30b] reported reactor production of I^{125}, and by 1960 Myers,[30d] Harper,[31] and others were using research quantities of the isotope in a number of applications. By 1961 a reactor production process had been developed sufficiently that ORNL could announce[32] the ready availability of this isotope; however, two years later, on Oct. 1, 1963, the AEC withdrew[33] from routine production of I^{125} and turned this operation over to private industry, in keeping with established policy.

Iodine-125 was originally desired as a replacement for I^{131} in medical diagnostic work. Some of the pertinent physical properties of the two radioisotopes are:

Isotope	Half-life, days	Electromagnetic radiations, Mev	Beta, Mev
I^{125}	57.4	0.0354, 0.0274	None
I^{131}	8.05	0.36, 0.64	0.33, 0.61

Myers and Vanderleeden[30d] and Harper et al.,[34a] pioneers in the use of I^{125}, have summarized its properties and advantages: (1) I^{125} gamma and X rays can be more efficiently detected by small, light scanning equipment; (2) compounds prepared with I^{125}, because of its longer half-life and lower energy radiations, have a longer shelf-life than those prepared with I^{131}; and (3) a patient to whom the compounds are administered receives less radiation because of the absence of beta radiation. Some of the tracer uses that have been reported include thyroid[35] and kidney scanning,[36] simultaneous study of individual portions of the two lungs,[37] differential diagnosis of steatorrhea[38] by labeling one compound with I^{125} and another with I^{131}, and biological studies[39] (e.g., in autoradiography of bacteria).

With lowered costs, larger scale uses become feasible, and both medical and industrial radiography sources are now being developed. Their usefulness—also due to the long half-live and low-energy, nearly monochromatic, electromagnetic radiation of the I^{125}—is shown by the excellent detail found in roentgenograms of human bones, for example, those published by Myers,[40] Söremark and coworkers,[41] and ORNL in the Fall issue of *Isotopes and Radiation Technology*,[42] and in a guinea pig skeleton shown in this issue (Fig. I-15). A description of an industrial source and good pictures made with it were also published recently.[43]

With further cost reduction, I^{125} sources for therapy are feasible.* Exposure to personnel is greatly decreased, and bone marrow of patients is shielded by the bone itself.

Iodine-125 with low I^{126} (13-day half-life) content is generally desired since I^{126} decays with beta emission (0.38 to 1.5 Mev) and with positron emission (0.46 and 1.1 Mev), and it also emits hard-gamma rays (0.65 to 1.4 Mev). The I^{126} content can be kept to a low value by using short-term irradiations, by using a low flux, or by allowing the shorter lived I^{126} to decay to a lower level. The method actually used depends primarily upon the production rate required. Some of the possibilities are covered in this report, and additional details

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*Personal communications to W. G. Myers, Ohio State: B. Pierquin, July 8, 1960, and Feb. 15, 1963; U. Henski (N. Y.), Dec. 20, 1960; J. Mallard (London), Aug. 2, 1962; and D. Lawrence (Palo Alto), June 28, 1963.

Fig. I-15 Radiograph of guinea pig made with a 1.96-curie I^{125} source. Time, 1.5 hr; distance, 10 in.; film, Du Pont Cronar with no intensifying screen.

will be presented in future issues of *Isotopes and Radiation Technology*.

I^{125} Production

Reactor Production. The most practical means at the present time for producing I^{125} is reactor irradiation of Xe^{124} (Fig. I-16). This reaction has been used in a batch process at ORNL[44] for the past two years to produce I^{125} in multicurie amounts, as well as at ANL[34] to product I^{125} for research use. The I^{126} content has varied from <1% up to about 15%, depending on irradiation time and flux.

The cost of I^{125} depends on many factors, including the I^{126} contamination allowable, the production rate, the production method used, the amortization of any costly equipment involved, and the cost of the target. Cost studies, based on operational experience and submitted to the AEC in July 1961, indicated that I^{125} with <15% I^{126} contamination could be produced readily in curie quantities for <\$1.00 per millicurie (exclusive of development costs and payments in lieu of profit, taxes, and insurance) by irradiating normal xenon in pressurized capsules.

For most applications, <1% I^{126} is desirable. Since the formation of I^{126} during I^{125} production increases with irradiation time and also appears to increase approximately as the square of the neutron flux, the percentage of it in the I^{125} product is minimized by using short-term irradiations or by using a lower flux. Since both of these measures reduce the yield, this effect is alleviated by using enriched xenon targets or large amounts of natural xenon and by using compressed gas targets to get more xenon in the irradiation field.

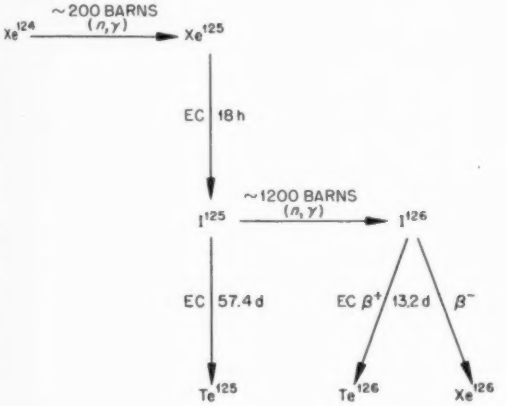

Fig. I-16 Production of I^{125} by irradiation of Xe^{124} (natural xenon containing 0.094% Xe^{124}).

In a typical batch run,[44] a 425-ml sample (STP) of natural-xenon gas contained in a 56-ml welded aluminum can at about 100 psi was irradiated in the ORR at a flux of 2×10^{14} neutrons/(cm^2)(sec) for 21 days. The resulting 1800 mc of I^{125}, containing about 15% I^{126}, was recovered from the container by washing it out in 0.1*N* NaOH solution, transforming to elemental iodine by appropriate chemistry, and distilling the carrier-free iodine into dilute sodium hydroxide. The recovery process is the same as that used for recovery from cyclotron targets.[44a] The unit full-recovery cost of this material, exclusive of the previously mentioned costs, was about \$1.20 per millicurie. However, irradiations at high pressures (up to 5000 psi) are feasible,[34a,b] and unit costs would be decreased with increasing pressure. For example, pressurizing the container to only 400 psi, for comparable flux and time, would either lower the production cost to ~\$0.30 per millicurie or would permit a decay of two months to yield product with ~1% I^{126} contaminant for ~\$1.20 per millicurie.

Cyclotron Production. Iodine-125 may also be produced in the cyclotron by the nuclear reactions $Te^{125}(p,n)I^{125}$, $Te^{124}(d,n)I^{125}$, and $Te^{125}(d,2n)I^{125}$. The I^{125} product from natural tellurium is very expensive and is highly contaminated with other iodine isotopes, although its production with minimum contamination is reported by Green and Martin.[30] They used 310 mg of 88% Te^{125} as TeO_2 in a sealed aluminum capsule and irradiated it with 16- to 18-Mev protons to favor the desired (p,n) reaction over the $(p,2n)$ reaction. The I^{126} (13-day) contamination was low because of the decreased amount of Te^{126} present in the enriched Te^{125}, and no other iodine radioisotopes were present in detectable quantities after one week of decay. The production rate of I^{125} was 1.8 mc/hr, a factor of 13 over that which would have been obtained with natural tellurium.

Enriched Xenon. Enriched Xe^{124} for the production of I^{125} has a number of advantages, particularly the tremendous reduction in the sizes and capacities of the irradiation facilities needed and the increase in product yield for a given facility under fixed conditions. For example, 10% Xe^{124} is enriched by a factor of 100 over natural xenon, so that the yield under a given set of conditions will be increased by a factor of 100.

At a continuous production rate of ~56 ml/day by thermal diffusion, the estimated[45] production costs for 10% Xe^{124} (including a one-year amortization of the plant) are ~\$5 per milliliter plus the cost of xenon and, for batch production of 5 ml of 10% Xe^{124} at a time, \$200 per milliliter plus the cost of xenon. In the continuous plant, depending on the type of thermal-diffusion system used (136 or 63 tubes, for example), the natural xenon required for the 56 ml/day of 10% Xe^{124} would cost between \$225 and \$640 per day, or between \$4 and \$12 per milliliter of 10% Xe^{124}. For large-scale continuous production, the "stripped" crude xenon possibly could be returned to normal channels[46] and, presumably, any use charge would not exceed 5% of the market price; this would add \$0.20 and \$0.60 per milliliter, respectively, to the cost of 10% Xe^{124}. Thus total production costs for the enriched xenon, adjusted to include the cost of raw, totally expended xenon, would lie between \$9 and \$18 per milliliter for continuous production and might be as high as ~\$220 per milliliter for batch production. With "borrowed" xenon the production costs would approximate \$5.50 per milliliter for continuous production and would be essentially unchanged for the batch method.

Thermal-diffusion columns for enriching Xe^{124} have been in operation for several months at ORNL, and the enriched product will be tested as target material for I^{125} production.*

Process Development. In theoretical studies at ORNL,[47] consideration has been given to three types of irradiation: (1) batch, (2) circulating loop, and (3) batch-operated loop. The results of calculations for both natural and enriched xenon indicate that all these methods except batch irradiation of natural xenon in the ORR can produce economic I^{125} in 5-curie amounts with an I^{126} content of <1%.

Considering enriched Xe^{124} as the target material, if present facilities, capsules, etc., were to be used, 15 ml of 10% Xe^{124} could be compressed into a standard ORR large capsule, and, after irradiation for 21 days at 10^{14} neutrons/(cm^2)(sec), the I^{125} product would be ~10 curies, produced at a cost of <\$500 plus the cost of the xenon (which would lie between ~\$150 and \$3300). However, the burnup of the Xe^{124} is <5% per run.† Conservatively, then, 10 runs, averaging >75% of the projected 10 curies per run, could easily be made by recovering the xenon. Thus the target cost would be between \$20 and \$450 per run, with a corresponding unit product cost of \$0.06 to \$0.10 per millicurie. It is apparent that increasing the pressure would further reduce the costs correspondingly.

For product with low I^{126} content, the pressure would be increased (to keep the yield high), and the irradiation time would be reduced (or decay time could be allowed), with an increase in cost to perhaps \$0.15 to \$0.25 per millicurie.

The use of loops for I^{125} production has a definite advantage in that the Xe^{125} intermediate can be removed from the irradiation zone, thereby greatly reducing the formation of I^{126}. Natural xenon can be used economically, but enriched Xe^{124} reduces the size of the loop significantly. This method of production, which has been under investigation by ORNL for some time, will be reported in a future issue of *Isotopes and Radiation Technology*.

Summary

The advantages of I^{125} over I^{131}, including more efficient detection of radiations, longer half-life, and lower energy of radiation, have led to a development program at ORNL aimed toward efficient and economical production of multicurie quantities of I^{125} by irradiation of Xe^{124}. A number of batch production runs have been made with targets of natural xenon under pressure. Unit full-recovery cost of 2-curie batches containing <15% I^{126} is ~\$1.00 per millicurie. Increased pressure and container size could reduce the cost to <\$0.10 per millicurie, and, with a circulating pressurized loop and enriched Xe^{124}, the cost could be further reduced to <\$0.05 per millicurie. Either by allowing this product to decay or by using short batch irradiations or a circulating loop, I^{125} containing <1% I^{126} can be prepared in 5-curie quantities for as little as \$0.30 per millicurie and in large quantities for <\$0.10 per millicurie.

Calcium-47 Production at ORNL

The method by which Ca^{47} is produced at ORNL is described here, together with a brief history of its production and other possible reactions for making it.* The $Ca^{46}(n,\gamma)Ca^{47}$ reaction is carried out in the ORR on targets that have been enriched in Ca^{46} in order to increase the specific activity of the reactor

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*The research quantities of enriched Xe^{124} obviously will not be produced for costs as low as those indicated for routine large-scale separations.

†At a flux of 2×10^{14} neutrons/(cm^2)(sec) and a three-week irradiation, with σ assumed to be 90 barns.

*An expanded version of this review will be issued as USAEC Report ORNL-IIC-2.

product.[48] The Ca^{46}-enriched material, ORNL's inventory of which, in October 1963, was 146 mg of >30%, 197 mg of 20 to 30%, and 1489 mg of <20%, all as $CaCO_3$, is also supplied to the Radiochemical Centre at Amersham, England, and to other groups for irradiation. As of October 1963, ORNL had distributed more than 700 mc of Ca^{47}, which currently sells for \$200 per millicurie.[49] This isotope is also available from Abbott Laboratories, Oak Ridge, Tenn.; the Radiochemical Centre, Amersham, England; and Zentralinstitut für Kernphysik, Dresden, East Germany.[50]

Calcium-47 is useful in biological and medical work, e.g., in the study of calcium metabolism and in the early diagnosis of primary and metastatic bone cancer. Because of its 4.5-day half-life,[51] it delivers a lower radiation dose than the commonly used 165-day Ca^{45}, and its 1.29-Mev gamma radiation[52] is easier to detect than the Ca^{45} 0.25-Mev beta. The medical use of this isotope in tracer studies was suggested in 1954 by Stickley[53] of the Medical Physics Group at Brookhaven National Laboratory, who estimated that the radiation from a product with a Ca^{47}/Ca^{45} ratio of 10 : 1 would be within tolerable limits for human patients and that an even lower ratio might be suitable for animal studies. A poll made by Abbott Laboratories of users of Ca^{45} indicated great interest in Ca^{47}, and potential world demand was estimated to be between 0.5 curie and 5 curies annually. At the Paris radioisotopes conference in 1957, Bauer[54] reported on work with Ca^{47} in human patients, and the great medical interest in this isotope is indicated by the participation of 21 persons from a dozen countries in the 1961 conference[55] sponsored by the International Atomic Energy Agency (IAEA) as well as by its extensive program[56] on medical uses.

Ca^{47} Production

Methods of producing Ca^{47} were discussed at meetings in 1956 attended by representatives of ORNL and Abbott Laboratories. In 1958 and 1959, meetings were held by the IAEA to discuss methods for producing medical-grade Ca^{47}. The most feasible of the potential methods appeared to be neutron activation of Ca^{46}, which requires enriched targets. Since natural calcium contains 600 times as much Ca^{44} as Ca^{46} (Table I-4) and since the Ca^{46} activation cross section is only twice that of Ca^{44}, the Ca^{47} product of the $Ca^{46}(n,\gamma)Ca^{47}$ reaction from a natural target is highly contaminated with Ca^{45} from the simultaneous $Ca^{44}(n,\gamma)Ca^{45}$ reaction. At the time of the 1958 IAEA meetings, calutron enrichment was marginal, so the IAEA allocated funds for preenriching the calutron feed in Ca^{46}. However, calutron enrichment improved, and the preenrichment studies were discontinued in 1959. Calutron enrichment has continued to improve, and an enrichment factor of 10,000 has now been achieved.[57]

At the 1959 meeting the IAEA consented to provide support to European investigators in-

Table I-4 ABUNDANCE OF CALCIUM ISOTOPES IN NATURAL CALCIUM*

Isotope	Abundance, %
Ca^{40}	96.97
Ca^{42}	0.64
Ca^{43}	0.145
Ca^{44}	2.06
Ca^{46}	0.0033
Ca^{48}	0.185

*From *Chart of the Nuclides*, 2d ed., prepared by the Institute of Radiochemistry, Nuclear Research Centre, Karlsruhe, 1961.

Table I-5 EFFECT OF TARGET Ca^{46} ENRICHMENT ON Ca^{47} CONTENT OF PRODUCT

Target			Irradiation		Ca^{47} produced, mc			Product Ca^{47}/Ca^{45} activity ratio
Ca, mg	Ca^{46}, %	Ca^{44}/Ca^{46} wt. ratio	Flux, neutrons/(cm^2)(sec)	Time, weeks	Total	Per mg of Ca	Per week at 10^{14} neutrons/(cm^2)(sec)	
8	0.7	9	2.5×10^{13}	3	0.4	0.05	10	1
8	0.7	9	2.0×10^{14}	3	4.4	0.55	13	1
4	0.55	7	2.8×10^{14}	3	2.5	0.63	13	3
4	2.0	5	2.8×10^{14}	3	8.5	2.1	13	3.5
8	2.0	5	1.8×10^{14}	1	3.9	0.5	14	1.5
2	18.5	0.3	1.8×10^{14}	3	22	11.0	11	24

terested in using Ca^{47}, and ORNL agreed to deliver Ca^{47} at a cost, including shipping and handling, of $300 per millicurie of activity, calculated at the time of shipment. The first shipments, totaling ~4 mc, were made in December 1959.

ORNL Process for Making Ca^{47}. The reaction $Ca^{46}(n,\gamma)Ca^{47}$ is used to produce 25 mc of product per month, with a Ca^{47}/Ca^{45} ratio greater than 20 : 1, by a 17-day irradiation of $Ca^{46}CO_3$ in the ORR at a flux of $>2 \times 10^{14}$ neutrons/(cm²)(sec). The targets have a Ca^{46}/Ca^{44} ratio of 6 : 1, with absolute abundance values of 0.46% Ca^{46} and 0.08% Ca^{44}. The $Ca^{46}CO_3$ calutron product, ~30% Ca^{46} and 5% Ca^{44}, is diluted to these values with 99.99% Ca^{40} (as $CaCO_3$) before irradiation to decrease loss during processing of the Ca^{46}, which is valued at $400 to $650 per milligram of contained calcium. The Ca^{40} is not activated appreciably during irradiation (Ca^{40} cross section, 0.2 barn; Ca^{41} half-life, $>10^5$ years) and does not introduce any problems in medical use. The irradiated target is dissolved in dilute HCl to give a product consisting of $CaCl_2$ in ~1*N* HCl. Typical products have chemical purities >99% and specific activities >150 mc per gram of calcium.

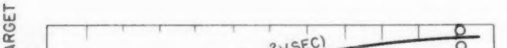

Fig. I-17 Effect of reactor irradiation time and flux of Ca^{47} production. Solid lines show actual yields, and the dotted line shows the theoretical yield at a flux of 2×10^{14} neutrons/(cm²)(sec); $\sigma = 0.25$ barn.

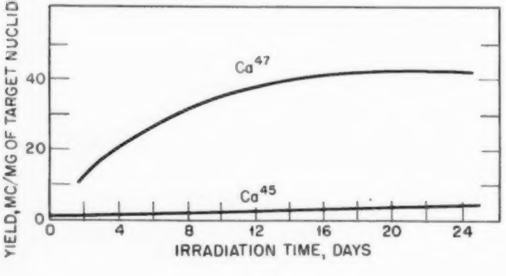

Fig. I-18 Effect of reactor irradiation time on production of Ca^{45} from Ca^{44} and Ca^{47} from Ca^{46}. Flux: 2×10^{14} neutrons/(cm²)(sec).

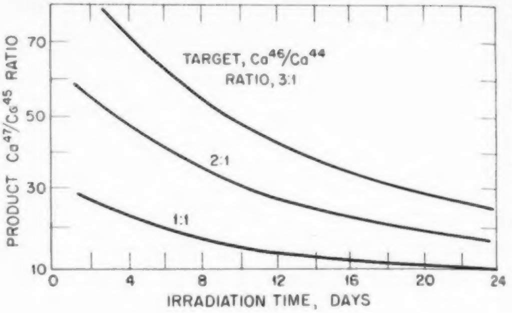

Fig. I-19 Effect of irradiation time and Ca^{46}/Ca^{44} ratio in target on Ca^{45} contamination of Ca^{47} reactor product.

The high Ca^{47}/Ca^{45} ratio in the reactor product is due primarily to the high Ca^{46} enrichment of the target material. In early work with a calutron product containing ~5% Ca^{46} and ~16% Ca^{44}, the reactor-product Ca^{47}/Ca^{45} ratio was 1 : 1. Increasing the Ca^{46} enrichment to 18.5% (6.0% Ca^{44}) gave a reactor product with a Ca^{47}/Ca^{45} ratio of 24 : 1 (Table I-5). For a fixed irradiation time, the Ca^{47} in the reactor product could also be increased by increasing the flux (Fig. I-17). On the other hand, increasing the irradiation time would not appreciably increase the Ca^{47} content (Fig. I-18) and would decrease the Ca^{47}/Ca^{45} ratio (Fig. I-19) because of the buildup of the longer half-life Ca^{45} impurity.

Other Nuclear Reactions Producing Ca^{47}. In addition to the $Ca^{46}(n,\gamma)Ca^{47}$ reaction, Ca^{47} can be produced in the cyclotron by bombardment of Ca^{48} or by transmutation of Ti^{50} (Table I-6). A number of these reactions are promising.

Ca^{48}(p,pn)Ca^{47}. The $Ca^{48}(p,pn)Ca^{47}$ cyclotron reaction is worthy of serious consideration and is being studied further because of the relative cheapness of the target, $10 to $25 per milligram of contained calcium for 70 to 95%

product. Enriched Ca^{48} target is prepared in the calutron, the enrichment now being 95% as compared with 50% in early separations.

The reaction has been demonstrated to be feasible in the ORNL 86-in. cyclotron with either a normal or a Ca^{48}-enriched target. The cross section is approximately 100 mb at 25 Mev (Fig. 3 in Ref. 58), and Ca^{45} contamination of the product from a Ca^{48}-enriched target is negligible. The reaction was not interesting in early work because of attenuation of the cyclotron beam by the capsule targets used and the inability to prepare adherent open-faced targets. However, adherent targets of normal CaO and CaF_2 have been made recently. Work is continuing on beam-current adjustment and target modifications in order to improve yields.

Table I-6 POTENTIAL REACTIONS FOR PRODUCTION OF Ca^{47}

Conversion of calcium isotopes
$Ca^{46}(n,\gamma)Ca^{47}$
$Ca^{48}(p,pn)Ca^{47}$
$Ca^{48}(n,2n)Ca^{47}$
$Ca^{48}(\gamma,n)Ca^{47}$
$Ca^{48}(d,dn)Ca^{47}$
[or $Ca^{48}(d,p2n)Ca^{47}$]

Transmutation
$Ti^{50}(n,\alpha)Ca^{47}$
$Ti^{50}(d,p\alpha)Ca^{47}$

$Ca^{48}(n,2n)Ca^{47}$. The $Ca^{48}(n,2n)Ca^{47}$ reaction has a cross section[58] of >1.5 barns. A cross section of 400 mb would be expected with the $d(H^3,\alpha)n$ reaction in Cockcroft-Walton or Van de Graaff machines, giving neutrons with energies of $\gg$14 Mev. With this reaction, since the Ca^{46} abundance is low compared with that of Ca^{48}, the Ca^{45} contamination would also be low.

$Ca^{48}(\gamma,n)Ca^{47}$. The $Ca^{48}(\gamma,n)Ca^{47}$ reaction with 70-Mev synchrotron-produced gamma rays could be useful if essentially complete absence of Ca^{45} is necessary. Voigt and others at Ames Laboratory[59] estimated that Ca^{47} with a Ca^{45} contamination of only 0.04% can be produced. The economics of the reaction has not been completely evaluated.

$Ca^{48}(d,dn)Ca^{47}$. The reaction $Ca^{48}(d,dn)Ca^{47}$ [or $Ca^{48}(d,p2n)Ca^{47}$] is not very attractive at present because of the very high deuteron energies required and the low peak cross section for the $(n,2n)$ reaction.

$Ti^{50}(n,\alpha)Ca^{47}$. The product of the $Ti^{50}(n,\alpha)Ca^{47}$ reaction on natural titanium in a cyclotron should have a high specific activity since the Ca^{47} would be separated from the titanium target. However, the activation cross section is low, 10 mb with 14.5-Mev neutrons,[60] and yields would be low. Also, since the target contains almost 80% Ti^{48} and the $Ti^{48}(n,\alpha)Ca^{45}$ reaction occurs simultaneously, Ca^{45} in the product would be high. Enrichment of the target from the normal 5% Ti^{50} to 85% would increase the yields considerably, and, since the Ti^{48} content would be reduced to ~10%, the Ca^{45} contamination would be reduced considerably. If the unconverted target could be completely recovered, very large targets could be considered capital items, making production by this transmutation reaction feasible.

$Ti^{50}(d,p\alpha)Ca^{47}$. The $Ti^{50}(d,p\alpha)Ca^{47}$ reaction has a peak at ~30 mb but at a rather high energy, 35 Mev. Hence this reaction is much less attractive than the (n,α) reaction.

Summary

Calcium-47 is produced by the $Ca^{46}(n,\gamma)Ca^{47}$ reaction in a high-flux reactor from target material electromagnetically enriched in Ca^{46}. The product, which sells for $200 per millicurie, has a Ca^{47}/Ca^{45} ratio of >20 : 1 and is made at a rate of 25 mc per month.

Stable Isotopes

Stable Isotopes from Fission Products

By R. S. Pressly

During fission and the subsequent decay processes, many stable and radioactive isotopes are formed. The fission-product radionuclides are discussed frequently,[61] but their stable congeners tend to be overlooked. However, some of these, because of their unusual isotope ratios, are worth considering as calutron charges (for example, the neodymium product mentioned below) in preparation of enriched stable isotopes.

The expected theoretical abundances of stable fission-product isotopes in two-year-old wastes

Table I-7 COMPARISON OF AMOUNTS OF STABLE FISSION-PRODUCT ISOTOPES AND NATURALLY OCCURRING ISOTOPES*

Isotope		Fission yield, %	Amount, %		Yield per 50 kilocuries Cs^{137}, g	Thermal-neutron cross section, barns
Z	A		In fission†	In nature		
32	Ge^{70}	0	0	20.55		
	Ge^{72}	1.5×10^{-5}	0.6	27.37		0.94
	Ge^{73}	1.0×10^{-4}	4.1	7.67		13.7
	Ge^{74}	3.0×10^{-4}	12.4	36.74		0.6
	Ge^{76}	2.0×10^{-3}	82.9	7.67		0.15 + 0.30
33	As^{75}	8.0×10^{-4}	100	100		
34	Se^{74}	0	0	0.87		
	Se^{76}	0	0	9.02		
	Se^{77}	9.1×10^{-3}	2.5	7.58		40
	Se^{78}	2.0×10^{-2}	5.6	23.52	3.0	0.4
	Se^{80}	8.0×10^{-2}	22.3	49.82	12	0.03 + 0.5
	Se^{82}	0.25	69.6	9.19	38	2.0
35	Br^{79}	3.7×10^{-2}	21.7	50.52	6	2.6 + 7.6
	Br^{81}	0.133	78.3	49.48	20	2.6
36	Kr^{78}	0	0	0.354		
	Kr^{80}	0	0	2.27		
	Kr^{82}	3.5×10^{-5}	0.09	11.56		45
	Kr^{83}	0.48	13.0	0	73	205
	Kr^{84}	1.1	29.9	56.90	167	0.1 + 0.06
	Kr^{86}	2.1	57.1	17.37	319	0.06
37	Rb^{85}	1.2	30.7	72.15	182	0.05 + 85
	Rb^{87}	2.7	69.3	27.85	410	0.14
38	Sr^{84}	0	0	0.56		
	Sr^{86}	2.8×10^{-5}	0.04	9.86		1.3
	Sr^{87}	0	0	7.02		
	Sr^{88}	3.7	100	82.56	562	0.005
39	Y^{89}	4.8	100	100	729	1.4
40	Zr^{90}	0	0	51.46		0.1
	Zr^{91}	5.9	23.7	11.23	896	2.0
	Zr^{92}	6.1	24.5	17.11	926	0.2
	Zr^{94}	6.5	26.1	17.40	987	0.07
	Zr^{96}	6.4	25.7	2.80	972	0.05
41	Nb^{93}	0	0	100		1.1
	Sn^{119}	0.01	13.7	8.58	1.5	
	Sn^{120}	0.01	13.7	32.97	1.5	0.001 + 0.03
	Sn^{122}	0.013	17.8	4.71	1.9	0.001 + 0.1
	Sn^{124}	0.02	27.4	5.98	3.0	0.75
51	Sb^{121}	0.014	48.2	57.25	2.1	5.7
	Sb^{123}	0.015	51.7	42.75	2.3	2.5 + 0.03 + 0.03
52	Te^{120}	0	0	0.089		
	Te^{122}	0	0	2.46		
	Te^{123}	0	0	0.87		
	Te^{124}	0	0	4.61		
	Te^{125}	0.015	0.6	6.99	2.3	1.5
	Te^{126}	0.10	3.8	18.71	15	0.07 + 0.7
	Te^{128}	0.5	19.1	31.79	76	0.016 + 0.14
	Te^{130}	2.0	76.5	34.49	304	
53	I^{127}	0.25	100	100	38	6.1
54	Xe^{124}	0	0	0.096		
	Xe^{126}	0	0	0.090		
	Xe^{128}	0	0	1.92		
	Xe^{129}	4×10^{-4}	0	26.44		45
	Xe^{130}	0	0	4.08		
	Xe^{131}	2.9	16.0	21.18	440	120
	Xe^{132}	4.4	24.3	26.89	668	? + 2
	Xe^{134}	7.6	42.0	10.44	1154	? + 2
	Xe^{136}	3.2	17.7	8.87	486	0.15
55	Cs^{133}	6.5	100	100	987	0.016 + 26
56	Ba^{130}	0	0	0.101		
	Ba^{132}	0	0	0.097		
	Ba^{134}	0	0	2.42		
	Ba^{135}	0	0	6.59		5.6
	Ba^{136}	6×10^{-3}	<0.1	7.81	<15	0.4
	Ba^{137}	5.9	50.4	11.82	896	
	Ba^{138}	5.8	49.6	71.66	880	0.68

42	Mo^{92}	0	0	15.86		
	Mo^{94}	0	0	9.12		
	Mo^{95}	6.4	27.1	15.70	972	13.4
	Mo^{96}	5.7×10^{-4}	0.02	16.50		1.0
	Mo^{97}	6.2	26.27	9.45	942	2.0
	Mo^{98}	5.9	25.0	23.75	896	0.13
	Mo^{100}	6.5	27.5	9.62	988	0.2
44	Ru^{96}	0	0	5.5		
	Ru^{98}	0	0	1.8		
	Ru^{99}	0	0	12.8		
	Ru^{100}	0	0	12.6		
	Ru^{101}	5.0	45.5	17.1	759	2.46
	Ru^{102}	4.2	38.2	31.7	638	1.2
	Ru^{104}	1.8	16.3	18.6	273	0.7
45	Rh^{103}	2.9	100	100	440	138.0
46	Pd^{102}	0	0	1.0		
	Pd^{104}	0	0	11.0		
	Pd^{105}	0.9	69.8	22.2	137	
	Pd^{106}	0.29	22.5	27.3	44	
	Pd^{108}	0.08	6.1	26.7	12	0.07 + 11
	Pd^{110}	0.02	1.6	11.8	3.0	? + 0.04
47	Ag^{107}	0	0	51.35		30
	Ag^{109}	0.028	100	48.65	4.2	2 + 82
48	Cd^{106}	0	0	1.215		
	Cd^{108}	0	0	0.875		
	Cd^{110}	2×10^{-7}	0.033	12.39		
	Cd^{111}	0.018	30.5	12.75	2.7	
	Cd^{112}	0.011	18.6	24.07	1.6	0.03 + ?
	Cd^{113}	0.01	16.9	12.26	1.5	25,000
	Cd^{114}	0.01	16.9	28.86	1.5	0.14 + 1.1
	Cd^{116}	0.01	16.9	7.58	1.5	1.4
50	Sn^{112}	0	0	0.95		
	Sn^{114}	0	0	0.65		
	Sn^{115}	0	0	0.34		
	Sn^{116}	0	0	14.24		
	Sn^{117}	0.010	13.7	7.57	1.5	
	Sn^{118}	0.01	13.7	24.04	1.5	
57	La^{138}	0	0	0.089		
	La^{139}	6.0	100	99.91	911	8.8
58	Ce^{136}	0	0	0.193		
	Ce^{138}	0	0	0.250		
	Ce^{140}	6.3	51.6	88.5	956	0.6
	Ce^{142}	5.9	48.4	11.07	896	1.8
59	Pr^{141}	6.0	100	100	911	11
60	Nd^{142}	0	0	27.13		
	Nd^{143}	6.2	27.8	12.20	1005	290
	Nd^{144}	6.1	27.4	23.87	927	4.8
	Nd^{145}	4.2	18.8	8.30	638	52
	Nd^{146}	3.3	14.8	17.18	501	9.8
	Nd^{148}	1.8	8.1	5.72	273	3.3
	Nd^{150}	0.74	3.1	5.60	112	
62	Sm^{144}	0	0	3.16		
	Sm^{147}	1.7	50.2	15.07	258	
	Sm^{148}	0	0	11.27		
	Sm^{149}	1.3	38.3	13.84	197	50,000
	Sm^{150}	0	0	7.47		
	Sm^{152}	0.3	8.8	26.63	46	150
	Sm^{154}	0.09	2.7	22.53	14	5.5
63	Eu^{151}	0	0	47.77		1400–7000
	Eu^{153}	0.15	100	52.23	23	480
64	Gd^{152}	0	0	0.20		
	Gd^{154}	0	0	2.15		
	Gd^{155}	0.013	36.4	14.73	1.9	70,000
	Gd^{156}	0.013	36.4	20.47	1.9	
	Gd^{157}	0.0074	20.7	15.68	1.1	160,000
	Gd^{158}	0.002	5.6	24.87	0.3	4
	Gd^{160}	3×10^{-4}	0.8	21.90		1.5
66	Dy^{156}	0	0	0.052		
	Dy^{158}	0	0	0.090		
	Dy^{160}	0	0	2.294		
	Dy^{161}	8×10^{-5}	0	18.88		
	Dy^{162}	0	0	25.53		
	Dy^{163}	0	0	24.97		
	Dy^{164}	0	0	28.18		

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*Taken in part from USAEC Report ORNL-2127, Part I, Vol. 1, J. O. Blomeke and M. F. Todd, Uranium-235 Fission-Product Production as a Function of Thermal Neutron Flux, Irradiation Time, and Decay Time. I. Atomic Concentrations and Gross Totals, Nov. 12, 1958.

†The percentage of a stable isotope of a fission-product element is based on the ratio of its fission yield to the sum of the fission yields of all isotopes of that element in the fission products.

Table I-8 NEODYMIUM FROM FISSION PRODUCTS*

Nd isotope	Natural abundance, %	Fission-product abundance, %	
		Calculated	Actual
142	27.1	0.0	0.0
143	12.2	27.8	27.7
144	23.9	27.4	26.5
145	8.3	18.8	19.1
146	17.2	14.8	14.9
148	5.7	8.1	8.3
150	5.6	3.1	3.5

*The origin of the fission products was long-irradiated natural uranium that had decayed about two years.[64]

from processing of fissioned uranium are shown in Table I-7. Corrections have been applied for those cases in which the half-life of the parent isotope is between six months and 10 years. However, no attempt has been made to correct for transmutative decay of radioisotopes to stable species or for burnup of stable isotopes since such corrections are functions of the operating time of the reactor and vary with total neutron dose (nvt); hence the percentages are only approximate. The number of grams of each isotope occurring with each 50,000 curies of Cs^{137} (a radioactive fission product recovered in large quantities[62]) is also shown. For example, for each 50,000 curies of Cs^{137} produced during fission, 182 g of Rb^{85} would also be produced. However, the actual amount remaining after a time in the reactor at a particular neutron flux would depend on Rb^{85} burnup as well as on decay of any other isotopes to Rb^{85}.

During the processing of radioactive fission products at ORNL,[63] 99.99% pure neodymium has been separated. Table I-8 compares the isotopic abundances of this material with those in so-called natural neodymium and with values calculated from fission-product yields. The agreement is remarkable.

Milking Systems: Status of the Art

By Margaret W. Greene, Robert F. Doering, and Manny Hillman*

The present status of milking systems used to obtain radioisotopes free from contamination by parent isotopes or other elements is reviewed here. Milking systems as means of separating a desired radioisotope from its parent and other contaminants have proved to be extremely useful. The milking system, or "generator," incorporates a relatively long-lived parent, fixed usually in a solid form, from which its shorter lived daughter can be separated as required. Thus relatively short-lived isotopes can be stored at the place of use. The system must, however, meet certain requirements in order to be of real value: (1) it must be simple to handle, operate, and shield; (2) it must yield rapidly a product of high purity in a chemical form suitable for use with little or no treatment; and (3) the parent material must remain in, or be reconverted readily to, a form with which the process can be repeated. Some milking systems are also described by Stang.[65]

The half-lives and principal radiations of nuclides discussed here are summarized in Table I-9.

Inorganic Sorbers

A relatively new and successful medium for preparing carrier-free Zr^{95} is unfused Vycor glass. Niobium-95 is adsorbed on the powder in a column, and 99% pure Zr^{95} is eluted with $10M$ HNO_3 (Ref. 66).

Practical systems using alumina have been developed at Brookhaven National Laboratory for use in milking I^{132} from Te^{132}, Tc^{99m} from Mo^{99}, and Ga^{68} from Ge^{68}. The I^{132} (Fig. I-20) and Tc^{99m} generators are made by adsorbing tellurite or molybdate on chromatographic alumina, and the products are eluted with $0.01M$ NH_4OH and $0.1M$ HNO_3, respectively. Both generators have been tested thoroughly in the United States and Europe.[67–74] The Ge^{68} generator, from which the daughter gallium is eluted rapidly by a neutral solution of ethylenediaminetetraacetic acid (EDTA)[75] replaces a more inconvenient and time-consuming extraction system.[76] In all three cases the purity of the product is high, with less than $10^{-3}\%$ radiocontamination and virtual absence of inactive carrier ions.

Organic Sorbers

For continuous preparation of carrier-free I^{132}, Münze developed a system in which Te^{132}

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*Brookhaven National Laboratory.

is precipitated on the hydrogen form of a cation-exchange resin saturated with stannous chloride, and the iodine daughter is eluted with water. The author states that the generator gives high yields and requires only simple manipulations.[77]

Practical working generators using cation-exchange resin have also been described for obtaining La^{140} from Ba^{140}, Ba^{137m} from Cs^{137}, and Y^{90} from Sr^{90}. When La^{140} is prepared, the parent Ba^{140} is sorbed on Dowex 1-X8 resin in carbonate form, and the daughter lanthanum is eluted with either $0.01M$ $(NH_4)_2CO_3$ or water.[78] The purity of the product is only fair since it contains about 0.2% Ba^{140}. Dowex 50 is used by Tracerlab, Inc., to separate Ba^{137m} from Cs^{137}, but no details of the chemical conditions are given in its reports.[79,80] The EDTA elution of Ba^{137m} from Cs^{137} sorbed on Dowex 50W has been described.[81] In the Y^{90} generator (Fig. I-21), which has been described in detail,[67,68] Sr^{90} is sorbed on Dowex 50, and the yttrium daughter is eluted with citric acid at *p*H 5.5. A decontamination factor of 10^{10} is obtainable.

In a recently developed Al^{28} generator,[82] Mg^{28} is sorbed on Dowex 50 cation-exchange resin, and Al^{28} is eluted by $1N$ NaOH solution containing 0.5 mg of inactive aluminum per milliliter. The yield is 30%, and 10 to 30 sec is required for elution. This isotope has the shortest half-live, 2.3 min, of any nuclide for which a useful generator has been developed.

Recently, two generators for Sr^{87m} have been developed independently. In one generator, available from Brookhaven National Laboratory, a citrate complex of Y^{87} is sorbed on Dowex 1-X8, and the Sr^{87m} is eluted with 0.005% citrate at *p*H 5.5. Less than 10^{-3}% radiocontamination and no inactive carrier ions are present.[83] In the second generator,[84] parent Y^{87} is sorbed on Amberlite IR-401 resin and is eluted with $(NH_4)_2CO_3$.

Nonsorption Generators

Two BNL-developed generators that do not rely on sorption are the early I^{132} generators. One of them relies on filtration for separation of the daughter iodine from precipitated parent tellurium dioxide.[85] The tellurium dioxide is dissolved in sodium hydroxide and is reprecipitated with acetic acid. The I^{132} that has grown into the solid tellurium dioxide dissolves in the caustic but is not precipitated by the acid. The resulting iodine solution is filtered off for use. In an earlier system[86] the parent Te^{132} was dissolved in a eutectic mixture of lithium and potassium chlorides. When the I^{132} that had grown in was to be milked off, the salt was melted and an air stream was bubbled through the molten salt to sweep out the iodine vapor, which was absorbed in a suitable solution. Yields of iodine were good when the vessel containing the molten mixture was glass; however, the mixture rapidly etched the glass, eventually causing it to break.

A distillation system was developed in England by Arnott and Peruma.[87] The parent tel-

Table I-9 HALF-LIVES AND PRINCIPAL RADIATIONS

Parent				Daughter			
		Radiation energy, Mev				Radiation energy, Mev	
Isotope	$t_{1/2}$	β	γ	Isotope	$t_{1/2}$	β	γ
Ba^{140}	12.8 days	1.02, 0.48	0.54, 0.16	La^{140}	40.2 hr	1.34, 1.10 0.42	1.6, 0.49 0.81, 0.33
Cs^{137}	30 years	0.52, 1.18		Ba^{137m}	2.6 min	IT	0.662
Sr^{90}	28 years	0.54		Y^{90}	64 hr	2.27	
Te^{132}	77 hr	0.22	0.23	I^{132}	2.3 hr	0.9, 2.12	0.78, 0.67
Mo^{99}	67 hr	1.23, 0.45	0.74	Tc^{99m}	6 hr	IT	0.142, 0.140
Ge^{68}	250 days	EC	X ray	Ga^{68}	68 min	1.88, 0.78 β^+	1.10
Mg^{28}	21.3 hr	0.45	1.35, 0.95 0.40, 0.032	Al^{28}	2.3 min	2.87	1.78
Y^{87}	80 hr	0.7 β^+	0.48	Sr^{87m}	2.8 hr	IT	0.39
Ti^{44}	10^3 years	EC	0.16	Sc^{44}	4.0 hr	1.47 β^+	2.54, 1.16
Sn^{113}	118 days	EC	0.26	In^{113m}	1.73 hr	IT	0.39
Ar^{42}	3.5 years	(3.5, 2.0)	(0.152)	K^{42}	12.4 hr	3.5, 2.0	1.5
Nb^{95}	35 days	0.16	0.76	Zr^{95}	65 days	0.40, 0.36	0.73, 0.76

lurium is left in contact with a reducing agent, hydroxylamine sulfate; iodine is removed, as required, by passing a stream of air through the parent solution. This generator can be recharged merely by adding fresh tellurium to the material already in the distillation flask. If the yield shows signs of falling below that expected, 0.5 ml of 20% H_2O_2 is introduced to suppress the reduction of iodine to iodide by the hydroxylamine.

In a system[88] described by Cook, Eakins, and Veall, I^{132} is allowed to grow, as the iodate, into a 10% H_2SO_4 solution of Te^{132} containing potassium permanganate. This solution is treated with 0.1N hydroxylamine sulfate to reduce the iodate to iodine, discharging the permanganate color.* Some iodine is also reduced to iodide, but it is reoxidized to iodine with a few drops of hydrogen peroxide. The generator is prepared for the next distillation by cooling and adding permanganate.

Other Systems

A German patent claims to separate I^{132} from Te^{132} and fission products by precipitating the tellurium and other metal ions with ammonia on a hygroscopic material that absorbs them. The iodine is then rinsed out with a sodium chloride solution or with hot water.[89]

Fig. I-21 Y^{90} generator.

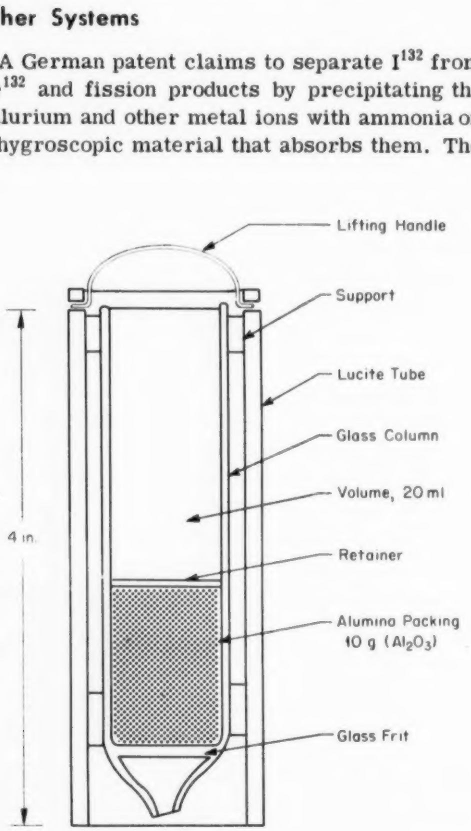

Fig. I-20 Iodine-132 generator.

Other useful generator systems that either are being developed or are under consideration are the pairs Ti^{44}-Sc^{44}, Sn^{113}-In^{113m}, and Ar^{42}-K^{42}. Some desirable characteristics of these systems are the marked differences in length of half-life between parent and daughter and the nature of the daughter radiation.

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Section

II

Isotopes and Radiation Technology

Isotope Technology Development

The applications of isotope technology to industry and to research may be conveniently divided into the following broad fields: (1) neutron activation analysis, (2) radiometric analysis, (3) tracer uses, (4) isotope measurement and control systems, (5) radiation absorptiometry, (6) nondestructive testing by radiography, and (7) ionization technology. The first four of these fields are currently emphasized in the development program of the AEC Division of Isotopes Development. In the first issue[1] of this quarterly, research on activation analysis and radiometric analysis was reviewed. In this issue of *Isotopes and Radiation Technology* the use of isotopes in measurement and control systems and the preparation of sources for use in nondestructive testing will be discussed, and in the next issue the use of isotopes as tracers will be emphasized.

Radioisotope Measurement and Control Systems

The AEC contracted with the National Industrial Conference Board in 1958 to survey the industrial uses of isotopes in the United States. The estimated annual savings made by almost 600 companies using isotopes, as determined by the Board, are summarized[2] in Table II-1. The table shows that the largest industrial use of isotopes is in gauging. This field has been well developed, and commercial instruments are available for measuring, for example, thickness of sheets and coatings, bulk and liquid density, liquid level, and water content of many materials. It is not within the scope of this review to discuss these commercial instruments; such details are available from the manufacturers. (The *Nucleonics* 1963/1964 Buyers' Guide[3] lists 28 domestic and 16 foreign companies manufacturing isotopic gauging equipment.)

Radioisotope gauges have the following important advantages over more conventional types of measurement equipment:

1. Located externally. The instrument is located outside the process equipment, and no contact is required with the material being measured. Conditions of temperature and pressure and the presence of corrosive materials do not affect measurements.

2. No sampling required. Since the measurement is made on the main process stream, no sampling tubes are necessary. If a sheet material is being measured, the process does not have to be stopped to remove samples.

3. No moving parts. All measurements are made electronically; therefore maintenance costs are low, feedback for process control is easy, and the overall stability of the system is high.

4. Insensitive to turbulence. In measurements of liquids, turbulence in pipes and ducts does not affect the measurement since the gauges are completely external to the process.

5. Clean. The chances of introducing contamination into a process during sampling are eliminated since the gauge is external, and no sampling is required. This high degree of cleanliness makes this type of measurement particularly attractive and safe for the food and drug industries.

Basic Principles. Isotopic gauges to measure thickness or density operate either by transmission or backscattering of radiation (Fig. II-1).

In a transmission type gauge (Fig. II-1*a*), the intensity of radiation (I) passing through the sample and reaching the detector is determined by the equation:

$$I = I_0 \, e^{-\mu \rho x}$$

where I_0 = the intensity of the original radiation
μ = the mass-absorption coefficient of the sample and depends on the type and energy of the source and on the atomic number of the sample
ρ = the density of the sample
x = the thickness

Thus a gauge of this type can measure either density at constant thickness or thickness at constant density.

The amount of radiation scattered by a sample (Fig. II-1*b*) increases with increasing thickness of the material up to a saturation value that depends on the type of radiation and the atomic number of the scattering material. With beta sources the saturation value is proportional to the square root of the atomic number and does not change much with beta-particle energy. With a gamma source the saturation value decreases with increasing atomic number.

Although the basic principles of radioactive gauges are simple, the practical design of gauges requires optimization of such variables as source type, source strength, detector type, and distance between the source and the detector. *Radioisotope Application Engineering*[6] discusses the design of gauges from both a theoretical and a practical viewpoint. Zumwalt[7] reviewed the theory for beta transmission-and-

Table II-1 ESTIMATED ANNUAL SAVINGS FROM RADIOISOTOPES DURING A 12-MONTH PERIOD IN 1957-1958

Industry	No. of companies	Net savings, thousands of dollars				
		Gauging	Radiography	Research	Manufacturing and processing	Total
Chemicals	38	1,247	75	2,325	803	4,450
Crude petroleum and natural gas	20	41	11	(42)*	810	820
Drugs	20			813	193	1,006
Electric and gas companies	4		5	7		12
Electrical machinery	16	172	10	101	12	295
Electronic components and accessories	19			782	8	820
Engineering and architectural services	59		402	1,743	3	2,148
Fabricated metal products	45		595	20		615
Food and kindred products	14	34		186	583	803
Machinery (except electrical)	26	10	248	531	206	995
Metal mining	4	253		13		266
Paper and allied products	93	2,826		(8)*		2,818
Petroleum refining and related industries	32	4,123	17	5,052	2,480	11,672
Plastics and synthetic resins	30	1,385				1,385
Primary metal industries	85	1,449	757	33		2,239
Rubber and miscellaneous products	26	2,845		(4)*		2,841
Stone, clay, and glass	20	551		289	34	874
Textile mill products	15	420				420
Tobacco manufacturers	†	2,955†		(50)*		2,905
Transportation equipment	30	279	692	751	7	1,729
Grand total		18,590	2,812	12,542	5,169	39,113

*Net deficit. Costs exceeded savings that could be measured during the census year.

†Because proprietary information is involved in data collected from tobacco companies, annual costs of equipment and facilities for gauging cannot be listed; however, these costs have been taken into account in figuring the net savings.

reflection gauges and used this theory to determine optimum conditions for the performance of these gauges. Kimball[8] described the use of isotopic gauges to solve four problems in the rayon industry and gave some practical suggestions to help others avoid difficulties when using these gauges. In two articles, Schurz[9] discussed, respectively, the practical and theoretical aspects of thickness measurements by radioisotopes. Dietzsch[10] discussed sensitivity and errors in both thickness and density measurements with the transmission method.

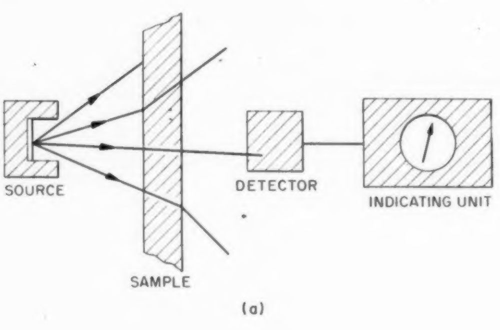

Fig. II-1 Schematic drawings of (*a*) transmission and (*b*) backscattering isotopic thickness or density gauges.[4,5]

Thickness Gauges. The use of radioisotope techniques for measuring and controlling thickness and density was well established by the time of the First International Conference on the Peaceful Uses of Atomic Energy at Geneva in 1955, and a number of papers were presented on the subject.[11] These techniques have a tremendous versatility; it makes little difference whether the material is metal, glass, paper, or plastic; these devices can measure them.[12–17] At the Midwestern Conference on the Industrial Uses of Isotopes in 1959, Aebersold[18] emphasized this extreme flexibility:

> Here I might point out something that management often misses. It is easy to see a profitable use in a given industry and fail to generalize, to see that the same principle can be applied in your own industry. A thickness gage that can control the thickness of rubber fabric can also control the thickness of soda cracker dough. A density gage controlling the ore content in a slurry can also be applied to processing noodle soup. Yet a food manufacturer might not be aware of such profitable possiblities.

Two thickness gauges can be combined[19] for measuring and controlling the thicknesses of coatings on paper or other materials; one gauge measures the uncoated feedstock, and the second measures the coated product. A simple computer calculates the difference between the two gauges to give the coating thickness. This radioisotope procedure not only is much faster than the traditional "sample-and-weigh" method but it is also continuous and nondestructive. The close control of tolerances greatly reduces off-specification scrap.

Backscattering procedures have been described in a number of recent papers on gauging;[20] Russian workers, in particular, are active in this field.[21–24] Recent development of bremsstrahlung and characteristic X-ray sources has added more variety to the possibilities for determining thicknesses.[25–28]

The savings to be realized from the use of radioisotopes in thickness gauges in industry come from the following factors:

1. Automation. The principal advantage of these gauges is that they lend themselves so easily to automation. As the detecting device measures the thickness, the instrument response can be incorporated in a servomechanism that automatically increases or decreases the spacings between the rollers to very close tolerances. Thus the response of the process to changed conditions is very rapid.

2. Lower consumption of raw materials. With other types of control, the manufacturer must set his thickness tolerances on the high side to meet minimum requirement at all times. Since the automatic radioisotope-actuated gauges permit closer tolerances through better control, much material can be saved.

3. Reduced scrap. The isotope gauge allows the equipment to get on stream rapidly since it is not necessary to cut and weigh control samples; scrap production is thereby greatly reduced.

4. Increased productivity. Because of the shorter startup time required to get product within tolerances, actual productive time is increased.

5. Reduced manpower. The fact that the gauges permit incorporation of automatic adjustments as an integral part of the quality-control measurements eliminates the need for personnel to make manual adjustments and to do tedious analyses.

Density Gauges. Obviously, the measurement of density[29] is related to the measurement of thickness. The first assumes a fixed thickness, and the second, a fixed density; therefore the equipment, at least in principle, is almost identical.

By far the largest single use of isotopic gauging equipment is in the tobacco industry to control the "firmness" of cigarettes.[30] Several thousand of these gauges are in continual use in the United States, where it is estimated that they save about $3 million a year (Table II-1). Density gauges are also quite satisfactory for detecting interfaces in pipeline flow,[31] and gauges of this type have now almost completely replaced soluble tracers injected at the interface.[32] The change in absorption of radiation from one liquid to another as the interface passes the detector can be used to signal the arrival of the second liquid.

An unusual application[33] of a density gauge is the measurement of the degree of polymerization of butadiene and styrene in the manufacture of synthetic rubber. As the two reactants polymerize, the density changes; the detector, having been calibrated in terms of percentage completion of the polymerization, can be incorporated in a control system to stop the reaction.

Isotopic density gauging has been used successfully in many different industries and with many materials,[6,34] for example, sugar solutions, slurries (of lime, clay, and metal ores), suspensions, brines, bases, acids (e.g., battery acid), black liquor (in the paper industry), condensed milk, ground corn, starch, tomato paste, foamed latex, synthetic rubber, gasoline, jet-engine fuels, and rock-wool blankets.

Level Gauges. A density gauge can also be used to determine the level of the contents of a container or to detect when the contents reach a certain level in a container being filled. The source is usually placed on one side of the vessel or container, and the detector, on the other. The source and the detector can be moved to locate a static level, or both can be set at a predetermined level to register the instant that the contents reach that level; in either case, the change in density introduced by the contents alters the radiation reaching the detector.[11d,11e,35]

Sometimes it is desirable to keep track of variations in liquid levels within a container. In this case the source can be placed vertically above or below the container, with the detector at the opposite end so that radiation readings show the liquid level.[36] The source can also be floated on the surface of the liquid, and the variation in radiation intensity, measured by a detector at the top, indicates the level. In the petroleum industry, liquid levels in refinery tanks and reaction vessels, as well as solid levels in storage bins and catalyst hoppers, are determined[11e] routinely by the movement of a Co^{60} "float."

Levels can also be determined with an isotope such as Cs^{137} and a backscattering technique, which depends on the fact that the amount of backscattering from the source is greater when it is below the level of the contents than when it is above.[37] A recent application of this type involved the detection of a propane-brine interface in a propane storage reservoir.[38] The interface was detected by the change in intensity of backscatter as the probe moved from the brine to the propane.

Gamma Backscatter Measurement Systems

Most of the isotopic measurement systems currently in use are based on the principles of beta backscattering or beta or gamma transmission; very few systems have been described that depend on gamma backscatter. In 1954 Hine and McCall[39] reported a study of the important factors in gamma backscatter, in which they considered the backscattering of gamma rays of various energies (0.280 Mev from Hg^{203}, 0.661 Mev from Cs^{137}, and 1.17 and 1.33 Mev from Co^{60}) from wood, aluminum, iron, and lead. They also investigated the effect of the thickness and geometry of the scattering material on the intensity of the scattered rays. In the same year Hayward and Hubbell[40] published the results of studies that they had made on Co^{60} gamma-ray backscattering from infinitely thick slabs of wood and steel wool. These two studies established many of the fundamental principles of the method.

In 1954 at Harwell, Putman and Solomon[41] used the scattering of the 0.085-Mev gamma ray from Tm^{170} to determine the amount of shale in coal. Putman used an experimental setup similar to that shown in Fig. II-1*b*, with a G-M tube as a detector for the scattered gamma rays; in later work[42] he eliminated the shielding between the detector and the source and used a scintillator crystal followed by a single-channel analyzer as a detector. The analyzer was set to select only the gamma rays of backscatter energies. Using this type of equipment, he built a portable analyzer that could determine the thickness of steel pipe up to 2 cm with an accuracy of ± 4%.

Contractors for the AEC Division of Isotopes Development have recently reported on several applications of gamma backscatter to isotopic measurement systems. Tolan, Bradshaw, Mears, McIntosh, and Hauck[43,44] at Lockheed have completed a study of the application of Compton backscatter to nondestructive testing. One part of this study was to develop sufficient information on gamma backscatter that an instrument design engineer could design a suitable instrument. Another part was to develop a gamma backscatter apparatus to detect flaws in heat-exchanger piping. Semmler, Brugger, and Rieke[45,46] at the Laboratories for Applied Sciences of the University of Chicago have investigated gamma-scattering density meters with particular reference to the determination of the density of coal in piles. Ely and Loos[47] have compared the use of gamma-ray backscatter with transmission for the evaluation of defects in logs and trees.

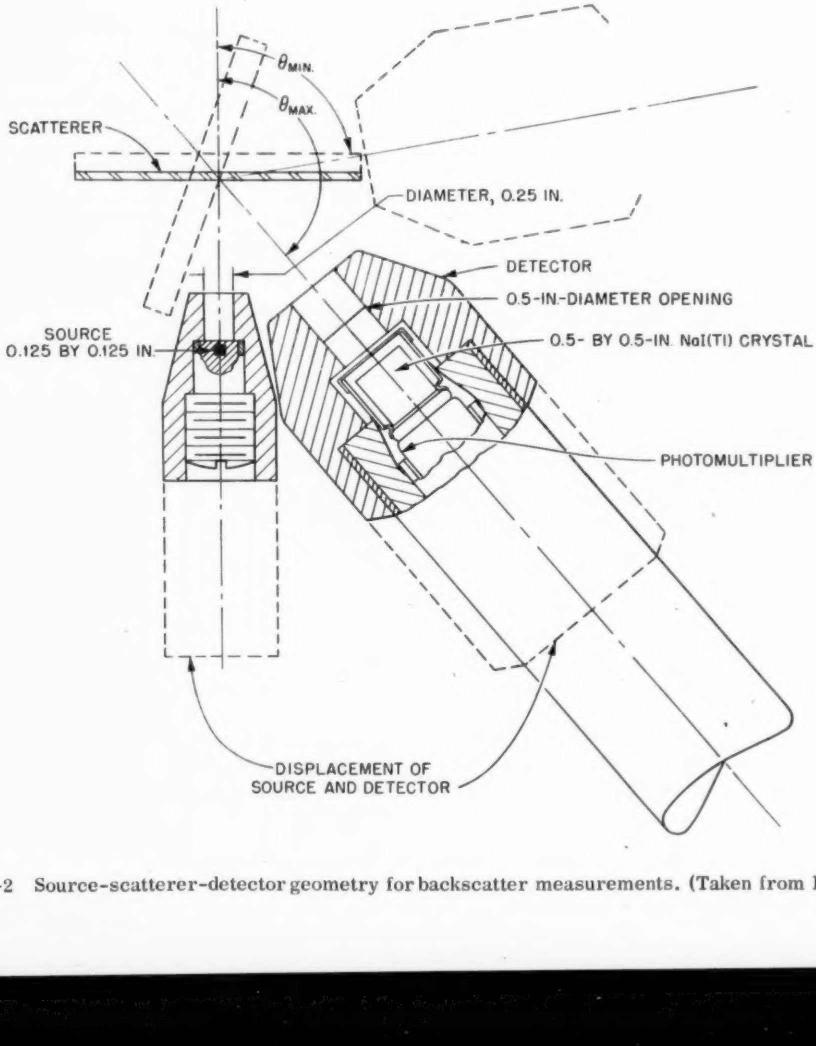

Fig. II-2 Source-scatterer-detector geometry for backscatter measurements. (Taken from Ref. 44.)

Design Data for Backscatter Gauges. One of the major goals in the research of Hauck and coworkers[43] at Lockheed was to obtain fundamental information on the backscatter process under a number of different experimental conditions so that the instrument design engineer would have more information available than had been published in the papers by Hine and McCall[39] and by Hayward and Hubbell.[40]

A modified optical spectrometer was used to make these measurements (Fig. II-2). The scattering materials were mounted on the grating table, which could be rotated about a vertical axis. The source was placed on a tray supported by the fixed post from which the light source for the spectrometer had been supported. The source could be moved along a line normal to, and intersecting, the axis of rotation of the specimen table. The detector was mounted on a tray supported by the movable post on which the telescope for the spectrometer had been mounted. This tray was also pointed toward the specimen so that the axis of the detector intersected the center of the specimen and the area of impingement of the source radiation.

The detector support post could be rotated about the specimen for independent adjustment of the scattering angle. A minimum value for the scattering angle was about 90° because angles less than this would not permit placing the source and the detector on the same side of the specimen. A maximum angle of 140° was established by the position of contact of the source and detector collimators. Independent adjustment of the angle of incidence of the radiation was permitted by rotating the specimen table.

Radiation energies from 0.084 to 0.660 Mev were evaluated in a standard geometry for a variety of materials ranging in density from plywood to lead. The initial result sought in the measurement was the relative quantity of radiation scattered at a given angle compared to the background radiation. This comparison established a signal-to-background ratio independent of source activity, and the ratio determined by the measurement was the basis for design criteria of a finished instrument.

Spectral scans of the signal plus background and background alone were not intended to be of the high quality of nuclear spectroscopy, but energy resolution of the spectral scans was sufficient to identify primary radiation transmitted directly through the collimator, backscattered radiation arising from the scattering material, and X radiation arising in and characteristic of the lead in the collimator near the crystal. Successive scans under identical conditions, except for the presence or absence of scattering material, can be compared with sufficient confidence to determine a signal-to-background ratio as a function of energy. The sources tested were Tm^{170} (0.084 Mev), Ce^{144} (0.134 Mev), Hg^{203} (0.280 Mev), Sn^{113} (0.390 Mev), and Cs^{137} (0.660 Mev).

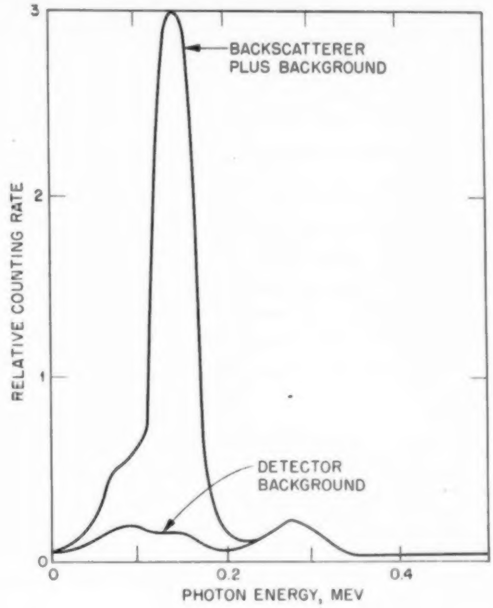

Fig. II-3 Backscatter spectrum of Hg^{203} (0.280 Mev). (Taken from Ref. 44.) Sample, 0.25-in. steel, NaI(Tl) detector, 0.5 in. in diameter and 0.5 in. long; scattering angle, 140°.

Of all the sources tested, Hg^{203} gave the most promising results (Fig. II-3). The curves show a clearly resolvable peak for the 0.280-Mev primary, a small X-ray component at 0.074 Mev, and a strong backscatter peak at 0.144 Mev. Because the signal-to-background ratio is diminished by direct and forward-scattered radiation transmitted through the collimator, the quantity of primary radiation is more than is desired. Thus the collimator thickness that was adequate for the radiations from Tm^{170} and Ce^{144} was not quite thick enough for those from Hg^{203}.

The scattering angle used in the backscatter measurement was investigated as a function of source radiation energy and scattering material. Each source was evaluated at different scattering angles to determine separately the response from wood, polyethylene, magnesium, aluminum, steel, and copper. Except for the changes expected from variation of electron density, the net signal increased in magnitude as the scattering angle increased, without passing through a maximum. Thin samples of 0.06 in. or less demonstrated a response maximum at an angle between 130 and 140°, but the change was very small. It was therefore concluded that a 140° scattering angle provides the maximum signal within the range investigated.

The sensitivity of response of different thicknesses of materials to the several radiation energies is shown in Fig. II-4. In Fig. II-4*a*, the family of curves representing backscatter signal as a function of thickness of material is plotted for the 0.084-Mev radiations from Tm^{170}. For the materials with higher density, such as lead, copper, and steel, photoelectric absorption of incident photons predominates, and even photons scattered by Compton interaction within the material are absorbed photoelectrically before escaping from the material.

Corresponding curves for Hg^{203} are shown in Fig. II-4*b*. With the high signal-to-background

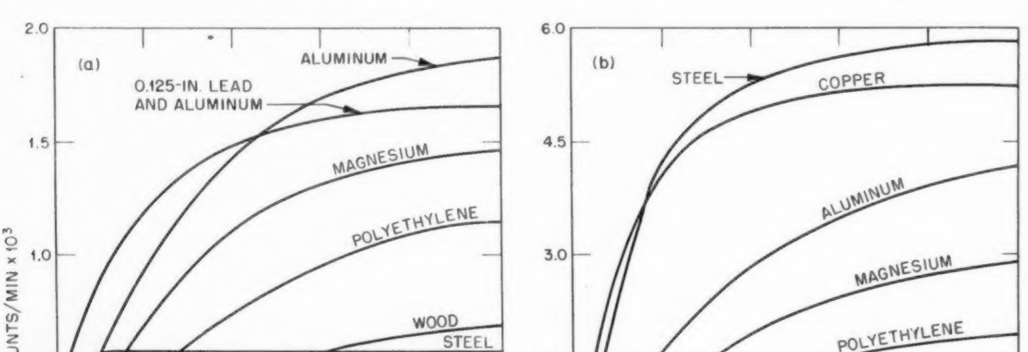

Fig. II-4 Sensitivity curves for (*a*) Tm^{170} (0.084 Mev); (*b*) Hg^{203} (0.280 Mev); (*c*) Sn^{113} (0.390 Mev); and (*d*) Cs^{137} (0.660 Mev). Scattering angle, 140°. (Taken from Ref. 44.)

ratio demonstrated with this source, all the numbers used in plotting the curves were reproducible within 1%; even this small error was probably the result of small changes in geometry rather than a statistical change in the quantity of backscatter. Saturation occurred in the steel and copper samples at a thickness of 0.4 in., but the slopes of the curves remained steep to 0.3 in. Thus the signal resulting from changes in thickness of steel permits a measure of thickness in the range 0.01 to 0.03 in. This range overlaps the useful, 0 to 0.06 in., range for the Tm^{170} source.

Other samples responded with good sensitivity to the radiations from Hg^{203}, but the response of aluminum, magnesium, polyethylene, and wood is better for the 0.084-Mev radiation from Tm^{170}.

Data for materials exposed to radiation from Sn^{113} are given in Fig. II-4*c*. The curves are strikingly similar to those for Hg^{203} (Fig. II-4*b*). Tin-113 would probably serve just as well as Hg^{203}, except that the signal-to-noise ratio is less for Sn^{113}, and currently available sources of Sn^{113} are 35 times as expensive as Hg^{203}. This latter factor far outweighs the advantage of longer life of Sn^{113}. (With a half-life of 127 days, Sn^{113} will last three times as long as Hg^{203}.)

The final source evaluated was Cs^{137} (Fig. II-4*d*). The signal-to-background ratio from this source was poor; consequently the numbers used for the curves were less reliable than desired. However, radiations from the Cs^{137} source possess a penetrating power not available in other sources, as evidenced by the increased thickness of steel and copper at which saturation occurs.

Pipewall Thickness Gauge. To test the effectiveness of gamma backscattering, equipment was built[43,44] to study the wall thickness of a heat-exchanger tube. The key component of this apparatus was the probe assembly (Fig. II-5), which could be inserted into the pipe and tested with both Sn^{113} and Hg^{203} sources. Figure II-6 shows some data obtained with this equipment. Scans were taken of the pipewall when the pipe was surrounded by water and by air, and satisfactory signals were obtained in both cases.

Density of Coal by Gamma Scattering. Semmler, Brugger, and Rieke[45,46] at the University of Chicago have made an extensive study of the design of gamma-scattering gauges. The following factors that may influence the operation of a gamma-scattering density gauge were investigated in detail: gamma source energy, source-detector separation, geometry of shield, and effect of chemical composition. The final report on this phase of the work[45] describes the effects of all these factors on the performance of density gauges.

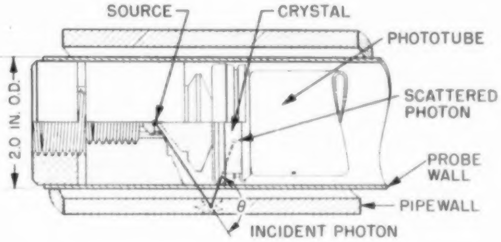

Fig. II-5 Prototype of pipewall-gauge probe. (Taken from Ref. 44.)

After this study was completed, the results were applied to a study of the determination of the density of coal piles at two steam plants that sell electricity to the AEC. In earlier work at these two plants, a commercial isotopic density gauge had been used (Nuclear-Chicago Corporation model P-20). Results were generally acceptable, but the calibration curves for the density of the coals at the two stations were not identical. The variation in response of the density gauge in different coals was thought to be caused by differences in the ash and possibly by the moisture contents. A second difficulty was that the calibration curve was not linear. A linear curve is advantageous for two reasons: (1) estimation of the calibration curve from the experimental data is easier and (2) calculation of inventory data is easier since each individual field reading must no longer be converted to a density before an average is computed (i.e., the field reading can be averaged directly, and this single value can then be converted to a density).

Two basic changes were made in the Nuclear-Chicago Corporation P-20 density gauge: (1) the source-detector separation was increased by $3\frac{11}{32}$ in. and (2) a cylindrical heavy-element filter was placed around the G-M tubes to absorb the low-energy gamma radiation. The source strength was also increased from 3.3 to 15.3 mc of Cs^{137} to maintain the counting rate at approximately the original value.

The heavy-element filter consisted of 0.005-in. copper, 0.020-in. cadmium, 0.003-in. lead, and 0.005-in. tantalum, with the copper foil next to the G-M tubes. The shield was 5 in. long and surrounded the sides and bottom of the bundle of three G-M tubes.

As a result of these two changes, the calibration curve is now linear in the region of interest, and the sensitivity (change in response per unit change in density) is also greater.

The intended effect of the heavy-metal filter was to strike a compromise between the errors introduced by variations in moisture content and in heavy-element (ash) concentration. The filter materially reduced the sensitivity to variations in heavy-element concentration; the only question was that of balancing the two sources of error. Since only one coal was used for the calibration, no conclusive comparison of the relative sensitivity to variations in composition was obtained. The degree of success of the particular choice made for the filter can be determined only by further comparison using different coals and moisture contents.

A troublesome and as yet unsolved problem in this work is the preparation of good standard samples to use in the calibration of the gauge. The scatter of the calibration points was still much greater than desired with either the modified or unmodified gauge. Undoubtedly some of the variation in the modified gauge response was due to mechanical difficulties; however, the unmodified gauge showed much the same variation. Some method other than tamping the coal and driving the tube by hand may be required to prepare more reliable samples. The hand-prepared barrels used here were prepared with reasonable care, but occasionally they still produced a questionable response.

Detection of Defects in Logs by Gamma Backscattering. Ely and Loos[47] at the Research Triangle Institute made a brief study of the use of gamma backscattering to detect defects in logs and growing trees. They were particularly interested in the possibilities of this method for determining the suitability of trees or logs in the manufacture of wood veneers.

It was concluded from their work that the backscatter technique is, in general, inferior to the transmission technique for the detection of interior defects in logs, trees, and utility poles.

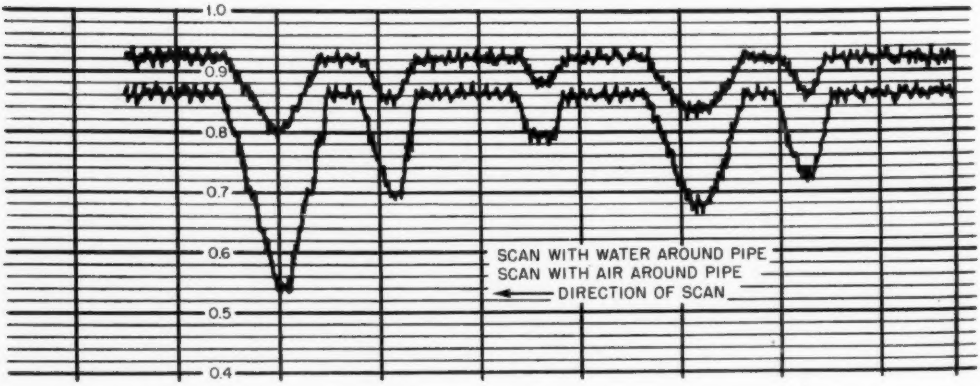

Fig. II-6 Pipewall scans with Hg^{203} source. (Taken from Ref. 44.)

When both sides of an object are accessible, the backscatter technique has an advantage only if the defect is 1 to 2 in. deep and the thickness of the wood is many times the thickness of the defect. In this case the defect appears to the transmitted beam as a small discontinuity, whereas, because of the shorter range of the backscatter beam, the discontinuity appears large to it. When only one side of an object is accessible and the backscattering technique must be used, Sn^{113} provides a greater sensitivity than Cs^{137}.

The backscatter technique does not appear to be promising for detecting internal defects of interest to the veneer industry, where the evaluation must be made on standing trees or green logs. About 45% of the defects that the veneer industry wishes to detect are knots < 0.5 in. in diameter—well below the capability of the backscattering technique—and in logs having diameters of 2 to 3 ft, also below the capability of the transmission method.

The backscattering technique is useful for detecting defects in 1- to 3-in.-thick planking. Some possible useful applications are the determination of dry rot in wooden ship hulls, the thickness of wooden floors in railroad boxcars, the thickness of wooden bridge floors, and the location of structural members behind sheet walls.

Density Gauge for Mass Flowmeter

Haffner[48] of the Illinois Institute of Technology Research Foundation (formerly Armour Research Foundation) and Genthe of the Badger Meter Manufacturing Company, Milwaukee, Wis., have developed a new apparatus for measuring the mass flow rate of a fluid in a pipe. This meter will handle multiphase fluids, is independent of external environment, and will not interfere with the flow pattern of the liquid. It can measure flow rates from 5 to 100 gal/min of liquids with densities of 1 to 1.45 g/ml, with an overall accuracy of ± 0.75%.

The mass flow rate is essentially the product of fluid density and fluid velocity, integrated in space over the cross-sectional area of the pipe. In order to measure these quantities (density and velocity) external to the flow, a section of the pipe was shaped like the letter S (Fig. II-7). This section, which was pivoted about its axis of symmetry, was connected with the remainder of the pipe by flexible couplings. Under the influence of fluid motion through it, the S-shaped section tended to rotate, and the rotation was counteracted by a spiral mechanical spring that was wound up by a torque motor controlled by a null-seeking servo. The angular rotation of the torque motor required to preserve the angular position of the S-shaped section was proportional to the reaction torque (T) of the fluid on the S-shaped section:

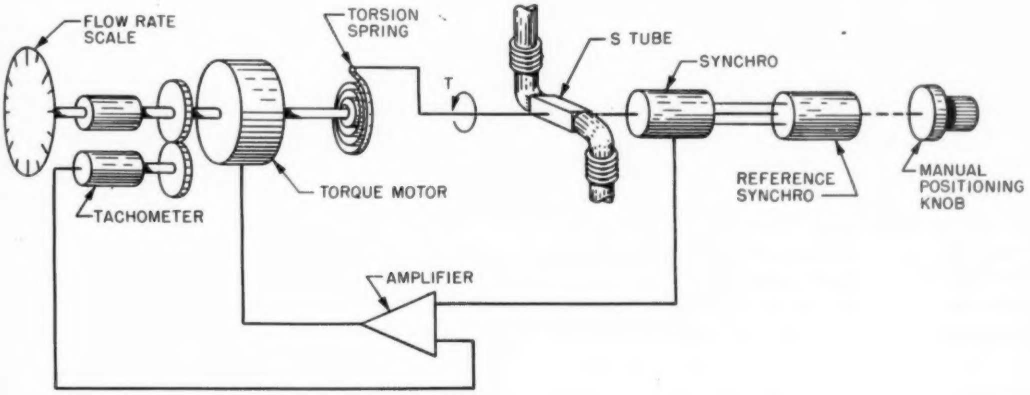

Fig. II-7 Schematic drawing of mass flowmeter. (Taken from Ref. 48.)

$$T = k\rho v^2$$

where ρ is the density of the fluid, v is the velocity of the fluid, and k is a constant (for any one gauge).

The density was measured independently by a gamma-transmission densitometer, which was designed to be independent of variations in line voltage, frequency shifts, photomultiplier-tube sensitivity, amplifier-gain changes, radioactive decay of the gamma sources, or radioactive materials entrained in the flow stream. This review will deal only with the densitometer part of the system since it is the part that uses isotopes. The original report[48] gives full details of the overall mass flow system.

Densitometer Description. The density was measured by comparing radiation from a standard source with that from another source after its rays were absorbed by passing through the S tube and the rotary wedge (Fig. II-8). Small circular windows permitted the gamma rays to pass across the center of the S tube. The center line of the S tube, the radiation path, and the axis of rotation of the S tube were mutually perpendicular. Flow rate and density must be measured as near the same point as possible to maintain accuracy. The servo loop controlled the angular position of the wedge so that the total attenuation of the fluid and the wedge remained constant. Since the same detector was used to measure radiation from the two sources, time must be shared between them. The dual radiation beams must be chopped so that one reaches the detector while the other is attenuated, and vice versa.

Various techniques were examined for chopping the radiation beams. One approach, a rotating shield with alternate sections open, was mechanically too cumbersome in practice. Another approach, radioactive sources mounted on vibrating reeds, was discarded because of the possibility of source damage. The final design used Hevimet shields mounted on the ends of electromagnetically driven reeds, with the sources held stationary behind the shields. In each case 60 cps was used, which meant that each source was on for $^1/_{120}$ sec at a time. The scintillation crystal was alternately exposed to radiation from one source transmitted through the flow stream and the calibrated wedge and to radiation from the reference source. Both sources were 9.7-curie Pm^{147} beta-excited X-ray sources.

The reeds themselves were made from strongly ferromagnetic stainless steel and designed so that stresses due to vibration were uniformly distributed over their lengths. Displacements were such that maximum stresses were well below the fatigue limits for the steel used. The vibrating reed itself did not make contact with any other member; thus wear problems did not exist. These facts combine to provide a simple and reliable radiation-sampling technique.

Vibration was sustained by applying an alternating magnetic field to the ferromagnetic material of the reed. If the reed is to be vibrated at line frequency (60 cps), it must have an undamped natural frequency of 60 cps to minimize input power necessary to sustain the vibrations.

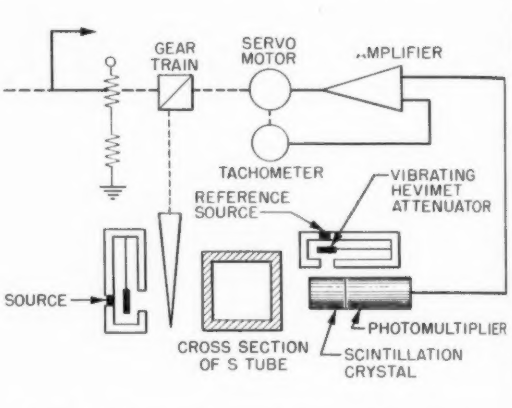

Fig. II-8 Schematic diagram of wedge densitometer. (Taken from Ref. 48.)

The scintillation crystal fluoresced under the effect of incoming gamma rays. Fluorescence in the crystal was converted by the photomultiplier tube into an electron current at the output of the tube. This current provided a net a-c error voltage, which was amplified and used in a servomotor to drive the calibrated wedge so as to balance out the error voltage. The position of the motor shaft was proportional to density.

Densitometer Calibration Procedure. Thirteen 1-gal samples of aqueous solutions of K_2CO_3, graduated in density from 0.9970 to 1.4615 g/ml, were prepared and used for static calibration of the densitometer. The densities of these solutions were determined in two ways: (1) by regulating K_2CO_3 and water concentration by weight during mixing and determining density from concentration tables and (2) by accurately weighing 100 ml of solution on a precision balance. All portions of the density servo were mounted in place on the flow-element housing. The reading of the density servo dials when the S tube was filled with water was noted. Then the K_2CO_3 solutions were added sequentially, and successive readings of the dials were noted for each solution density. From these data a calibration factor relating dial reading and density was derived. This factor was used in the calculation of the overall mass flow constant for the test model.

Densitometer Performance. The performance of the densitometer servo was then tested with a sequence of solutions with densities between 0.9970 and 1.4615 g/ml. The measurement made by the servo was in agreement with the calibration standard within ±0.2% over most of the range and within ±0.33% over the entire range. This is equivalent to about ±0.003 unit of specific gravity or metric density. The random nature of the error indicates that the precision of the density standard was probably about the same as that of the servo measurement.

Readings were made for each solution after a period of several minutes had elapsed to allow the heavily damped servo to reach quiescence. At quiescence the readout dials moved randomly through small angles (±2°), and readings were the average values at each quiescent point.

Determination of Silage Density by Gamma-Ray Attenuation

Wilkins, James, and Menear[49] of the U. S. Department of Agriculture, Agricultural Research Center, Beltsville, Md., have been using a radioisotope gauge since 1960 to determine the density of silage. An accurate determination of silage density is important in silo design because, as silage settles, its density increases; this change causes a buildup of unequal pressures in the silo, which can sometimes rupture the walls. Changes in density are also important in studying the quality of low-moisture silage.

Several methods have been used to determine the density of silage in a silo.[49] One of the first methods adopted was the "layer method," which consisted in removing a measurable volume of silage and weighing it. From the known volume and weight, the density could be calculated. This method was subject to error because of decompression in the silage when weight was removed.

In the "surface sample method," samples of known volume were removed with a suitable tool, and spot densities could be calculated. Decompression must be considered.

A considerable amount of research has been done with a horizontal core sampling method. Although special silos with access openings were required to obtain a horizontal core, this method had some decided advantages over the two methods mentioned above. Decompression was not a problem, and this method showed a variation in the density throughout the mass; however, changes in the shear value of the silage can introduce errors in density. All the methods discussed so far have the disadvantage of being "one-shot" methods and therefore allow only one density measurement at a specific location in the silage.

Figure II-9 shows a diagram of the device used to measure density in a precast concrete stave silo 12 ft in diameter and 25 ft high. When the silo was empty, a 3-in. steel pipe was mounted vertically in its center to provide a passage for the source carrier during the test. Originally it was intended to measure only the direct energy emerging radially from the silo. Because of the high densities, it was necessary to measure both direct and scattered energy.

Cesium-137 was chosen as the source because it has a long half-life (27 years) and is a monochromatic emitter of gamma rays at approximately 0.66 Mev. One curie of Cs^{137} was put in a Lucite wafer, which was then placed between two lead cylinders. This source container was designed to give a collimated beam of activity emerging radially. When the source

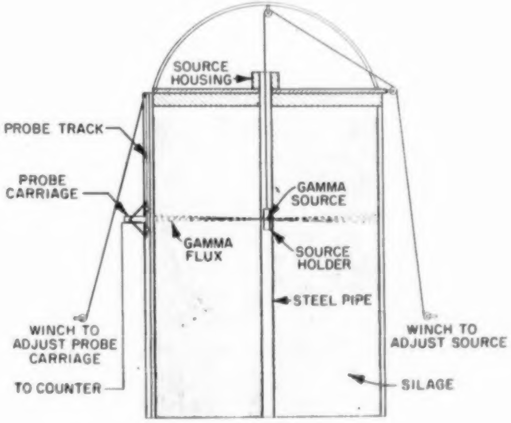

Fig. II-9 Location of source in silo. (Taken from Ref. 49.)

was not being used, it was locked in a lead shield.

The radiation detector was a 2-in.-diameter NaI(Tl) crystal with a photomultiplier tube and preamplifier. This probe was shielded with 0.5 in. of lead so that the background signal would be lower. A track on the outer surface of the silo guided the carriage as it was moved up or down. Originally the track could be hung on any one of four hangers around the periphery of the silo, but, to make the system more versatile, a monorail was installed to make it possible to leave the track at any location. The heights of both detector and source were controlled remotely by cables attached to a winch, which allowed separate movement of probe or source and also allowed synchronous movement of the probe and the source when desired. The signal from the preamplifier of the photomultiplier tube was transmitted to a pulse-height analyzer.

A typical curve obtained from wilted alfalfa hay silage is shown in Fig. II-10. The average density from this curve was 31.9 lb/cu ft, which compared favorably with the average density of 31.7 lb/cu ft by the weight method. This technique is excellent for detecting small changes in densities of silage in a silo, and repeated measurements can be made at any location. Nevertheless, the following complicating factors cannot be overlooked: (1) the radiation measuring equipment is expensive, (2) some knowledge of physics is essential, (3) interpretation of results may be slightly more difficult than with other techniques, and (4) only an average bulk density of the silage between the source and the detector is given. Although Wilkins and his coworkers feel that these are drawbacks, they intend to continue their research, not only to improve the method but also to learn more about factors influencing the density of silage in a silo.

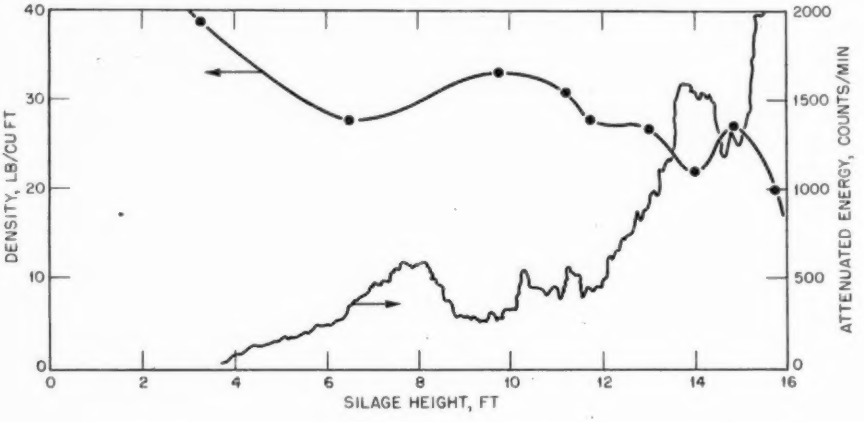

Fig. II-10 Change in silage density and attenuated energy with silage height.

High-Altitude Atmospheric Density Gauge

Ziegler[50] and coworkers at Parametrics, Inc., report the development of a successful beta-scatter gauge for determining atmospheric density at altitudes between 45,000 and 140,000 ft. This work was sponsored jointly by the AEC Division of Isotopes Development and the U. S. Weather Bureau.

The first part of the work was a detailed theoretical and experimental study to determine whether a beta-attenuation (transmission) gauge or a beta-scatter gauge would be more suitable for the determination of densities at high altitude. The results of this study are summarized in Fig. II-11, which compares the source strengths required for minimum errors in a density determination for an attenuation gauge (with C^{14}) and for a scatter gauge (with Kr^{85}). The forward-scatter configuration was superior to the attenuation gauge for the higher altitudes. At 140,000 ft, for example, the attenuation gauge required approximately ten times as much activity for equal accuracy. The crossover (equal activities) occurred at 67,000 ft. Below this altitude the attenuation gauge would be superior in terms of source strength

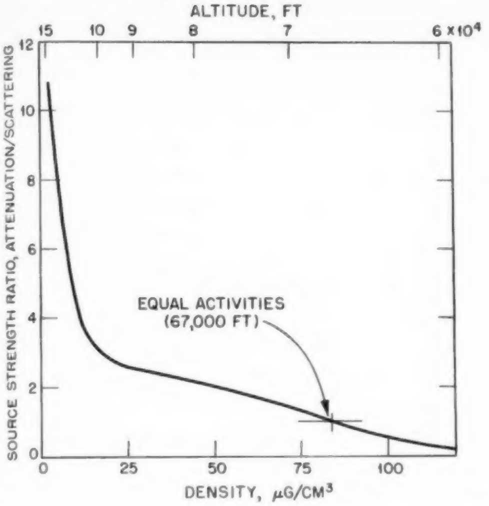

Fig. II-11 Source strength ratios required to give equal accuracies with attenuation and scatter gauges. (Taken from Ref. 50.)

requirements. The scatter gauge therefore was more suitable for the altitude range of interest (80,000 to 140,000 ft). The scatter gauge, which is optimized at 140,000 ft, would be usable to altitudes below 60,000 ft. The range of usefulness is thus greater than 80,000 ft.

The counting rate data for the optimized scatter gauge indicated that a source strength of a few millicuries of Kr^{85} would be required to achieve statistical accuracy of 1% in density determination in a stationary measurement lasting 1 min at an altitude of 140,000 ft, where a 1-sq in. detector area is assumed. Since activity requirements are inversely proportional to the useful detector area, a detector with an area of 2 sq in. would require half this activity.

The block diagram of the density sonde that was constructed and flight tested is shown in Fig. II-12. The detectors were of the scintillation type and consisted of anthracene crystal packages (with 0.0005-in.-thick aluminum windows) mounted on 1-in.-diameter photomultiplier tubes. A single high-voltage supply (1300 volts) was used to power both detectors. The "signal" detector was mounted so that it responded to atmospherically scattered beta rays originating from the source. The "background" detector was shielded from the source and therefore was not exposed. The responses of the two units to natural and cosmic backgrounds were the same. Pulses from the detectors were fed to pulse amplifiers, the outputs of which were coupled to wave shapers (single-shot multivibrators) of fixed sensitivity. By adjustment of the amplifier gain by the insertion of a potentiometer in the amplifier circuit, the sensitivities could be set to correspond to identical locations on the Kr^{85} beta spectrum. Amplifier gain was sufficient to give an operating point located on a sensitivity plateau, indicating that substantially all the beta spectrum was being detected.

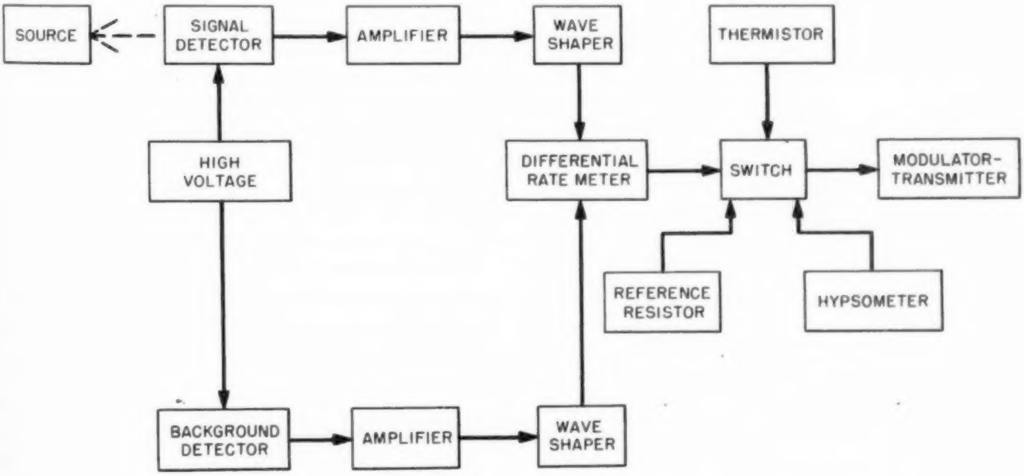

Fig. II-12 Block diagram of density sonde. (Taken from Ref. 50.)

Under these circumstances a plot of counting rate against detector voltage also showed a plateau, with 1300 volts at its midpoint. Thus very stable operation was achieved. The wave shapers were coupled to a difference rate meter that produced a voltage directly proportional to the net counting rate. The standard T-435 A/AMT-4B modulator-transmitter (frequency, 1680 Mc) was used. In normal use this transmitter is designed so that resistance variation (e.g., a thermistor) determines the repetition rate at which modulation is effected. The device may, however, also be voltage controlled by use of a suitable buffer stage. A switch, operated by a spring-driven clock mechanism, connected the rate-meter output, thermistor, hypsometer, and reference resistor to the modulator in cyclic sequence so that density, temperature, and pressure data were transmitted.

The complete sonde weighed 6 lb, including batteries. The Kr^{85} source strength was approximately 8 mc. The most useful and accurate range extended from approximately 240 to 2.8 $\mu g/cm^3$, corresponding to an altitude range of 45,000 to 140,000 ft, where the accuracy was better than 1%. Data can be taken from sea level to 150,000 ft, but measurements below 45,000 ft and greater than 150,000 ft are not so accurate.

Two density sondes were built and flown as required by the contract. On the first flight the mechanical clock used to switch from one sensor to another failed, and no useful data were obtained. On the second flight the sonde and transmitter functioned satisfactorily, but the ground receiver failed about 1 hr after launching; thus no useful data were obtained above 60,000 ft. However, sufficient data were obtained to illustrate system performance.

Air Force personnel calculated altitudes from pressure and temperature data by the usual method, which combines the use of the ideal gas law and the hydrostatic equation. A plot of the altitude-density profile thus obtained is shown in Fig. II-13. Flight of three more units will be undertaken as part of another contract.

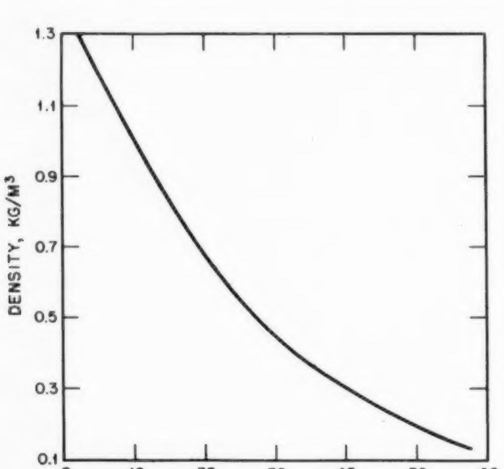

Fig. II-13 Change in density with altitude. (Taken from Ref. 50.)

Summary

Commercial radioisotope measurement systems have been developed for use in many applications, such as bulk density, liquid density, thickness of sheets, and firmness of cigarettes. The basic principles of these instruments are reviewed briefly, and the following new developments in the use of gamma backscatter gauges are described: (1) a gauge for measuring the thickness of heat-exchanger tubing by inserting a probe in the inside of the pipe; (2) a modification of a commercial coal density gauge to give it a more linear response and to make it less sensitive to the composition of the coal; and (3) the detection of defects in logs by gamma backscattering. A double-beam gamma density gauge developed for use in a mass flowmeter and gauges for measurement of density of silage and of the atmosphere from 45,000 to 150,000 ft are also described.

Radioisotope Sources for Radiography

By R. S. Pressly

X rays in the energy range from 0.005 to 0.1 Mev play an important role in the growing use of radiation in research and technology. Their principal areas of application are density and thickness gauging, industrial and medical radiography, and analysis by absorptiometry and fluorescent excitation techniques. The need for radiation in this energy range is currently covered by conventional X-ray machines. However, because these machines are not completely portable and require a source of electric power for operation, many investigations have been carried out to replace or supplement them with radioisotope sources. Two types of such sources are currently under investigation:[51] low-energy gamma-emitting isotopes and bremsstrahlung sources.

Low-Energy Gamma Emitters

Very few long-lived isotopes emit low-energy gamma rays, and, because of the complex decay schemes of most of them, they are of limited use when a narrow photon energy distribution is needed. Isotopes such as I^{125} and Fe^{55}, however, which decay by electron capture alone, give off monoenergetic photons. These isotopes have recently been prepared as effective low-energy gamma sources at ORNL. The work with Fe^{55} sources is described here, and the preparation of I^{125} sources is described in Section I.

Iron-55 Portable X-Ray Source. Iron-55, with a 2.94-year half-life, decays by electron capture to manganese, which emits a 0.0059-Mev X ray. It is prepared by neutron irradiation of enriched Fe^{54}. This isotope has been especially useful for determining sulfur in petroleum[52] because of the high absorptivity of sulfur for this low-energy photon. Electron-capture isotopes such as Cs^{131} and W^{181} are also useful for X-ray-fluorescence analyses,[53] but they can neither be prepared with high specific activities nor fabricated into point sources.

A source was made by spreading Fe^{55} oxide evenly over a circular area of a platinum strip by evaporating a nitric acid solution of $Fe(NO_3)_3$ and thermally decomposing the nitrate to the oxide. The oxide was reduced to metallic iron with hydrogen at 800°C, and the platinum strip was heated until the platinum on the surface started to melt and the metallic iron became coated with a thin layer of platinum. The area of platinum containing the radioactivity was cut from the platinum strip and rolled into a thin sheet. Repeated rolling of the platinum increased the radiation from the source because the iron was spread more uniformly and the thickness of the platinum coating layer was reduced. After a disk was cut, the area of the source was ~ 2.4 cm^2, and the radiation, as measured by a thin-wall ionization chamber, was 800 mr/hr at 1.5 in. from the center of the source.

Radiographs of flowers were made to illustrate that very small differences in densities can be clearly shown (Fig. II-14). No image intensifiers were used. A helium atmosphere blanketed the flowers to decrease the absorption and scattering of low-energy photons by the surrounding atmosphere, and the exposure time was adjusted to give the best radiographs. These pictures are comparable to similar photographs taken with a low-voltage X-ray machine.[54] The same techniques can also be used to radiograph watermarks in stamps and valuable papers.

Fig. II-14 Radiograph of pansies and leaves made with an Fe^{55} source in a helium atmosphere. Time, 4.5 hr. Film, Polaroid 3000 X.

Bremsstrahlung Sources

Radioisotope X-ray sources can also be made which use the bremsstrahlung produced when beta rays strike a target. When high-energy beta particles impinge upon a target, they lose their energy by radiative and ionization processes, most of the energy being dissipated in outer shell ionization of the target material. However, some of the energy of the electron goes into production of bremsstrahlung and characteristic X rays following inner shell ionization. The energy distribution and intensity of the total X-ray field depend on the nature of the target, the electron energies, and the strength of the beta source.

Several studies have been made recently on the preparation and use of bremsstrahlung sources. Brech of Jarrell-Ash Company made a brief study of the use of Pm^{147} in analytical fluorescence spectroscopy.[55] Bersin and co-workers at Tracerlab, Inc., studied the use of Kr^{85} clathrates with seven different targets[56] chosen to give a band of K_α energies between 0.01 and 0.1 Mev. Reiffel,[57] Filosofo,[58] and Ezop[59] at the Armour Research Foundation (now the Illinois Institute of Technology Research Foundation), who have been especially active in this field, have concentrated their attention on the beta-emitting fission products Pm^{147}, Kr^{85}, and Sr^{90} Y^{90}. They used tin and lead targets in most of their studies.

Promethium-147 X-Ray Sources. Promethium-147, a fission product now available in kilocurie quantities,[60] decays by emission of a 0.223-Mev beta ray to Sm^{147} with a 2.6-year half-life. There is no known stable isotope of promethium. Ezop[59] made a radiograph of acceptable quality of a human hand in 40 sec using a 250-curie Pm^{147} source 9 in. from Polaroid 3000 X film.

Sources at ORNL were fabricated by compressing high-purity promethium oxide into thin

Fig. II-15 Radiograph of a hand made with a 1000-curie Pm^{147} source at 16 in. Time, 5 sec. Film, Polaroid 3000 X.

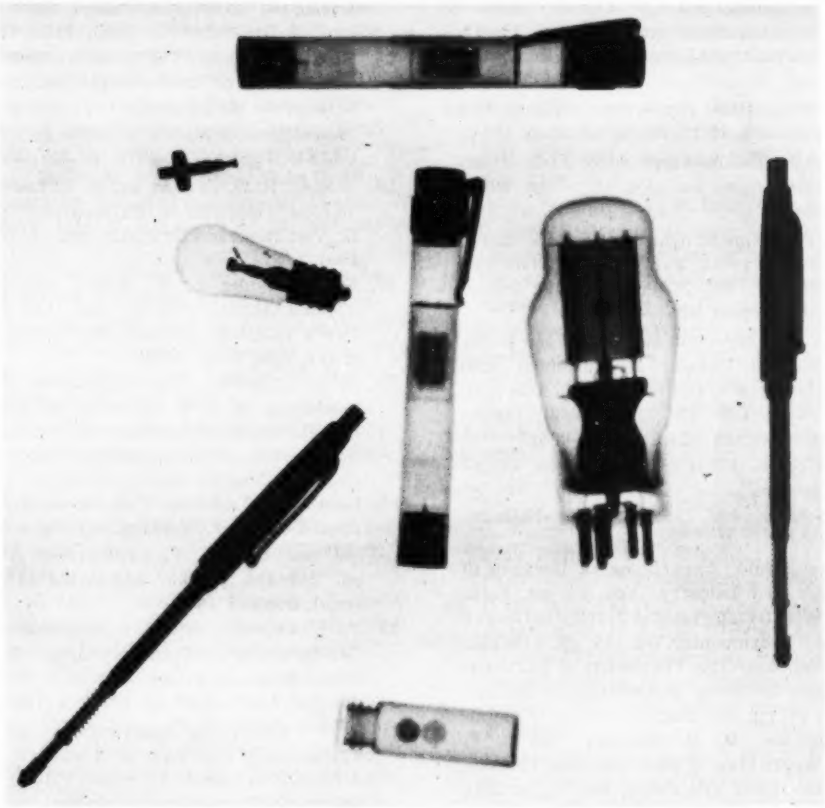

Fig. II-16 Radiograph of metal and plastic objects made with a 1000-curie Pm^{147} source at 16 in. Time, 10 sec. Film, Polaroid 3000 X.

pellets and encapsulating the pellets in aluminum source holders. One thousand curies of promethium formed a cylindrical pellet 2 cm in diameter by 1.5 mm thick. A window thickness of 0.015 in. in the source holder allowed 94% transmission of the 0.038-Mev photons.

The X radiation from the encapsulated 1000-curie Pm^{147} at 16 in., as measured by an ionization chamber, was 1 r/hr. Radiographs were taken of the human hand (Fig. II-15) with Polaroid 3000 X film and of certain objects to show how fine structures and density differences can be revealed (Fig. II-16).

The possibilities of the technique of using beta radioactivity for the production of secondary X rays have been shown. In particular, Pm^{147}, because of its long half-life, low-energy beta emission, and freedom from decay products that produce higher energy photons, is well suited for portable X-ray use.

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Section

III

Isotopes and Radiation Technology

AEC Program on Low-Dose Irradiation Processing of Food*

By Kevin G. Shea†

The Atomic Energy Commission program for the study of radiation pasteurization of food is described here.

Spoilage by microorganisms is the main cause of rapid deterioration of perishable foods, and, by decreasing the number of bacteria, ionizing radiation at low dose levels delays multiplication of bacteria and thus inhibits food damage. Hence the irradiation process, in combination with refrigeration, is potentially useful in extending the marketing life of foods for shipment to remote areas. Related applications include disinfestation of grains, sprout inhibition of potatoes and other vegetables, and control of insects for quarantine purposes. The related U. S. Army radiation-sterilization program is aimed at making a product that can be stored for months without refrigeration. Such a product is particularly suited to military logistics requirements but may ultimately have civilian applications as well.

In the radiation-pasteurization program, emphasis has been on a small number of selected food products that promise early technical feasibility and market acceptance. It is intended to define the processes for these products sufficiently to allow refinement and a final market test. Ultimately, the decision on success or failure will be made by industry in the market place.

The discovery of radioactivity by Becquerel in 1895 was followed closely by the idea of destroying microorganisms by this penetrating new force, with Prescott reporting[1] radiation effects on fungi in 1904. However, nuclear technology has only recently advanced to the point that large-scale use of radioactive materials has become feasible. The results of an AEC-sponsored survey[2] published in 1951 indicated that a successful radiation process for food would improve the nutritional and public health aspects of the American diet, and the AEC supported research in this field as early as 1950.

The first major effort in this field was by the Quartermaster Corps of the U. S. Army in 1954, with a program to develop sterilized food with a reasonable shelf life at room temperature. They have also supported some work on less-than-sterilizing doses.[3] The Office of the Surgeon General of the U. S. Army was, and still is, responsible for determining the wholesomeness of food in the Army experimental irradiation program. In a continuing broad research and development effort instituted in universities, nonprofit institutions, and private industry, more than 100 foods have been subjected to microbiological, chemical, and physiological studies; organoleptic properties of irradiated foods, packaging, dosimetry, and related matters have also been investigated.

The Army plans included construction of a research plant for food irradiation at the U. S. Army's Natick Laboratories, Natick, Mass., headquarters for the Quartermaster Corps Research and Development Program. This irradiation facility, built under AEC supervision, has a 1.1-megacurie cobalt source as well as an 18-kw variable-energy linear accelerator capable of providing electrons up to 24 Mev.

In March 1960 the United States Food Irradiation Program was realigned, and the AEC assumed the responsibility for the low-dose, or pasteurization, applications. This area suited

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the AEC objective of pioneering atomic energy developments that show special promise for industrial participation. The Division of Isotopes Development has the responsibility for source design and development and for determining optimum variables for product irradiation, and the Division of Biology and Medicine has the responsibility for the food wholesomeness aspects of the program. In December 1960 the American Institute of Biological Sciences established a committee of scientists to consult with the Commission's Division of Biology and Medicine and the Division of Isotopes Development on irradiation processing of foods. Eight meetings have been held at six-month intervals, the first in August 1960.

Close liaison is maintained by the AEC with the Army's Quartermaster Corps and with the Office of the Surgeon General. AEC personnel are members of the Interdepartmental Committee on Radiation Preservation of Foods, which provides channels of communication, through its constituent members, with the U. S. Departments of Agriculture, Defense, and Interior, the Food and Drug Administration, and other interested government agencies. The importance of radiation processing of foods in the Atoms-for-Peace Program has encouraged cooperation and maintenance of close liaison with other countries interested in this activity. The AEC participated in the Paris meetings of the Study Group on Food Irradiation of the Organization for Economic Cooperation and Development, and participating European nations have been kept fully aware of progress and plans. Contact has been maintained with other international organizations such as the International Atomic Energy Agency, World Health Organization, Food and Agriculture Organization, and Euratom.

AEC Program

Significant progress has been made in the technology necessary to extend the refrigerated shelf life of fruits and marine products by irradiation, and a commercial process has been outlined for several food items. The refrigerated shelf life of these products has been extended up to five times normal, with radiation doses lower than originally anticipated. Based on previous process evaluations in this[4] and other programs (Fig. III-1), the studies have been extended to include shipping and storage studies with a mobile Co^{60} unit located in California. Irradiation does not improve the quality of either fruit or marine products, and the preirradiation condition and handling of the food are therefore important. Contractors doing work sponsored by the Division of Isotopes Development are listed in Table III-1, part A.

Economics. Results now available from an economic analysis, being made by the U. S. Department of Agriculture,[5] of the process as applied to fruit are promising. Food industry representatives feel that radiation processing will increase the production and marketing value of selected fresh fruits and vegetables but will not change the output and sales volume of canned, frozen, and other processed forms of the same commodity. The next phase of the study will determine the cost of the irradiation process compared to existing treatments and the probable impact of the method on the prices, suppliers, and market structures involved.

The Bureau of Commercial Fisheries of the U. S. Department of the Interior is conducting a two-phase study, on the results of which an educational program will be based. The first phase will determine the potential impact of radiation pasteurization of marine products on market suppliers, structures, and losses. The second phase will be a highly qualitative study among consumers of fish and shellfish to determine the kind of educational program required for most successful marketing of irradiated seafood. This will involve a study of attitudes, information, misinformation, and psychological images related to fish in general and irradiated fish in particular.

Process. The program effort has been directed to the two classes of perishable foods, i.e., seafood and fruit, selected as showing the most promise, with evaluation of the product by Massachusetts Institute of Technology (MIT)[6] and Stanford Research Institute.[7] The seafood includes soft-shell clams, haddock, shrimp, crab, flounder, and certain fresh-water fish. The fruits include strawberries, citrus, tomatoes, peaches, cherries, figs, mangoes, and papayas.

Marine Products. Work[8] done at the Bureau of Commercial Fisheries Laboratories, Gloucester, Mass., shows that the refrigerated shelf life of fresh haddock and clams can be tripled with doses of 250,000 and 450,000 rads, respectively. The quality of the fish prior to

Fig. III-1 Effect of gamma radiation on Shasta strawberries, 1963. (*a*) Unirradiated berries. (*b*) Berries irradiated with 200 kr. (*c*) Unirradiated berries after eight days at 41°F. (*d*) Irradiated berries after eight days at 41°F.

irradiation greatly affects the shelf life of the irradiated product, with fish one day old keeping for 30 days but fish seven days old at the time of irradiation keeping for only 11 additional days.

Work done at MIT indicates that a dose of 250,000 rads extends the refrigerated storage life of haddock fillets seven to eight days at 43 to 46°F and 14 days at 32 to 33°F. For clams treated at the same level, the additional storage life is 16 days at 43 to 46°F and 21 days at 32 to 33°F.

At the Bureau of Commercial Fisheries Laboratories in Seattle, the shelf life of Pacific crab meat treated with 200,000 rads and held at 33°F was extended from a normal seven to 35 days. Petrale sole fillets treated with 400,000 rads responded similarly, their storage life being about twice as long at 33°F as at 42°F.

At Louisiana State University, freshly caught shrimp, when properly handled, have had an iced storage life of 14 to 16 days. Treatment with 50,000 to 100,000 rads doubled this time. Melanosis or "black spot," a discoloration that occurs in shrimp from the action of phenol oxidases, can be controlled by low doses, about 50,000 rads.

Fruits and Vegetables. Most of the work with fruit has been done at the Davis branch of the University of California.[9] Fruits that naturally have a short storage and market life will probably be of most interest for irradiation. Strawberries, sweet cherries, figs, oranges, papayas, and pineapples, and possibly peaches and nectarines, show the most promise to date. Strawberries and sweet cherries have been preserved with doses of 200,000 to 250,000 rads.

At the University of Florida in Gainesville, work has started on irradiation of tomatoes and citrus products. Michigan State University has initiated work on midwestern fruits, including cherries.

Table III-1 CONTRACTS IN EFFECT IN THE AEC PROGRAM ON LOW-DOSE RADIATION PASTEURIZATION OF FOOD

	Subject of study	Contractor
	A. Division of Isotopes Development	
Fruit	Gamma-radiation treatment for the control of postharvest diseases of fresh fruits and vegetables	U. S. Department of Agriculture, Agricultural Marketing Service, Chicago
	Radiation technology in conjunction with postharvest procedures as a means of extending the shelf life of fruits and vegetables	University of California, Davis
	Radiation pasteurization of foods	University of Michigan, Ann Arbor; also Michigan State University and Bureau of Commercial Fisheries
	Effect of low-level irradiation on the preservation of food products	University of Florida, Agricultural Experiment Station, Gainesville
	Irradiation of fruits and vegetables with AECL mobile irradiator	U. S. Department of Agriculture, Fresno, Calif.
Fish	Radiation preservation of fishery products	U. S. Department of Interior, Bureau of Commercial Fisheries, Gloucester, Mass.
	Application of radiation-pasteurization processes to Pacific crab and flounder	U. S. Department of Interior, Technological Laboratory, Seattle, Wash.
	Extension of storage life and growth of type E *Clostridium botulinum* in radiation-pasteurized marine products	Massachusetts Institute of Technology, Department of Nutrition, Food Science and Technology, Cambridge
	Radiation pasteurization of shrimp	Louisiana State University, Baton Rouge
	Effects of irradiation on the microbial flora surviving radiation pasteurization of shrimp	Oregon State University, Department of Food Science and Technology, Corvallis
	Radiation pasteurization of foods	University of Michigan, Ann Arbor; also Michigan State University and Bureau of Commercial Fisheries
Market evaluation	Marketing feasibility study of radiation-pasteurized fresh strawberries, peaches, tomatoes, grapes, oranges, and grapefruit	U. S. Department of Agriculture, Economic Research Service
	Study of attitudes and reactions of potential consumers of irradiated fishery products	U. S. Department of Interior, Bureau of Commercial Fisheries, Gloucester, Mass.
	Current status and potential of irradiation to prevent potato sprouting	Western Nuclear Corp., Idaho Falls, Idaho
Petition	Preparation and submission of petition on safe use of radiation on citrus fruits and marine products	Hazleton Laboratories, Falls Church, Va.
Packaging	Extraction study of packaging materials with irradiated foods	Hazleton Laboratories, Falls Church, Va.
Design and construction		Brookhaven National Laboratory
	B. Division of Biology and Medicine	
	Biochemistry of radiation and fruit maturation	University of California College of Agriculture, Davis
	Effect of ionizing radiation on resistance, germination, and toxin synthesis of *Clostridium botulinum* spores types A, B, and E	University of Michigan, Ann Arbor
	Low-level radiation preservation of fishery products	Fish and Wildlife Service Technological Laboratory, Gloucester, Mass.
	Radiation of heme proteins	University of California, Berkeley
	Study of the basic microbiological and biochemical factors involved in radiation preservation of marine products	University of Washington, Seattle
	Wholesomeness evaluation of radiation-substerilized foods (protein availability studies on irradiated soft-shell clams, haddock, crab, and flounder and two-year chronic studies on soft-shell clams)	University of Illinois, Urbana
	Wholesomeness investigation of radiation-pasteurized foods in the AEC food irradiation program: substerilized strawberries	Industrial Bio-Test Laboratories, Northbrook, Ill.

The University of Hawaii will investigate tropical fruits such as papayas, pineapples, and mangoes. Radiation treatment of Hawaiian fruits to meet U. S. mainland quarantine requirements is a logical application of the process, especially for control of the mango seed weevil. Equally important is investigation of shelf-life extension by control of plant pathogens. Fruits such as avocados, for which a chemical treatment is now approved, might be better treated by radiation.

Irradiators. Conceptual designs for a grain irradiator,[10] an on-ship irradiator,[11] and a multipurpose irradiator (fruit disinfestation and shelf-life extension) have been completed. Construction of the Marine Products Development Irradiator (MPDI), located at Gloucester, Mass., has been started and is scheduled for completion in late summer 1964. This $600,000 facility will operate on a near-commercial scale, processing marine products at rates of up to 1 ton/hr. It will be operated as part of the research and development program conducted for the AEC by the Technological Laboratory of the Bureau of Commercial Fisheries, U. S. Department of the Interior.

The engineering design of a Co^{60} mobile irradiator has also been started. This irradiator will be used, particularly on the West Coast, for large-scale studies of fruit processing.

Packaging. The Continental Can Company, Inc., recently completed a survey of packaging requirements[12] for radiation-pasteurized foods. Its report concludes that these requirements can be met by current technology and materials. The study, which involved the package engineering requirements of five fruit and five marine products, is based on interviews with specialists and on a literature review. Container materials, container design, and specific packaging methods are reviewed; areas of future research are recommended; and future required research, subsequent to Food and Drug Administration (FDA) approval, is outlined. As a result of this survey, a contract was let with Hazleton Laboratories to determine the effects of radiation on the selected packaging materials in order to evaluate their utility and safety. Ultimately, petitions will be prepared and submitted to FDA for approval of the selected packaging materials for their intended use.

Wholesomeness. The U. S. Army early established a research program[13] to determine the wholesomeness of radiation-processed food. The two-year tests in which irradiated foods are being fed are supported by in vivo and in vitro studies on growth, reproduction, hematology, histopathology, toxicity, carcinogenicity, and induced radioactivity. Twenty-one different foods, irradiated at sterilizing levels of 2.79 or 5.58 Mrads and stored for at least three months at room temperature, are incorporated into a basal diet as 37% of the solids. Statistical analysis of the final histopathological examinations will be completed shortly. The Army results, where applicable, are being extrapolated for use in the low-dose program.

The AEC Division of Biology and Medicine has a program to determine the wholesomeness of other irradiated foods of interest. The Division's existing contracts are listed in Table III-1, part B. A literature analysis[14] made by MIT to define the wholesomeness of marine products indicates areas where research is required. Data derived from U. S. Army studies at 3 and 6 Mrads may be extrapolated to the same class of foods for pasteurizing doses. However, further work on protein availability is required in such cases. This document, plus other recent information, was used to establish the research and development program needed to evaluate the nutritional quality and wholesomeness of the diet.

Two-year feeding studies in which soft-shelled clams are fed to selected subjects are under way, and similar tests are planned for strawberries. Protein availability studies on haddock, crab, flounder, and clams may lead to clearance of these food items for use.

A program has been initiated at MIT and Oregon State University in microbiology to determine the incidence of the various putrefactive anaerobes, including *Clostridium botulinum* type E, and the effects of low-dose irradiation on their growth and toxin production. In preliminary experiments at MIT, *Clostridium* counts were reduced significantly by low doses of irradiation and remained below 100 per gram at both 0 and 6°C, even after three months' storage. This is a rather encouraging result from the public health standpoint.

Acceptability. Subsequent to laboratory taste testing, the acceptability of the product after it has been processed and handled should be determined on a large scale. Preliminary tests

Table III-2 U. S. FOOD AND DRUG ADMINISTRATION REGULATIONS AND RELATED ACTIONS

Subject	Filed by	Petition
Gamma radiation for food processing, FAP 890	Department of the Army, Quartermaster Research and Engineering Command, Natick, Mass.	On Feb. 15, 1963, a regulation was issued in the *Federal Register* that radiation by Co^{60} may be used to sterilize bacon in coated cans by irradiation to an absorbed dose of 4.5 to 5.6 Mrads (33 FR 1465)
Gamma radiation for wheat treatment	Lloyd E. Brownell et al.	A regulation was issued in the *Federal Register* on Aug. 21, 1963, to include the following: gamma radiation from sources with maximum energy not to exceed 2.2 Mev, to provide an absorbed dose from 20,000 to 50,000 rads, may be safely used for irradiation of wheat and wheat products for control of insect infestation (28 FR 9208)
Electron-beam radiation for food processing	General Electric Company, Milwaukee, Wis.	On Aug. 30, 1963, a regulation was issued in the *Federal Register* to permit irradiation of canned bacon by an electron accelerator producing a beam of electrons at energy levels not exceeding 5.0 Mev (28 FR 9526)
Notice of filing of petition regarding food additive gamma radiation, FAP 1132, *Federal Register*, June 6, 1963	Department of the Army, Quartermaster Research and Engineering Command, Natick, Mass.	A regulation was proposed for use of gamma radiation at energy levels of not more than 2.2 Mev to inhibit sprout development in white potatoes with an absorbed dose of 5000 to 10,000 rads (28 FR 5588)
Notice of filing of petition regarding food additive, FAP 1205, *Federal Register*, Aug. 23, 1963	Department of the Army, Quartermaster Research and Engineering Command, Natick, Mass.	A regulation was proposed to provide for the safe use of electron-beam radiation with a maximum energy of 10 Mev and a dose of 4.5 Mrads for radiation preservation of canned bacon (28 FR 9329)
Letter, FDA to AEC, dated Aug. 26, 1963, requesting an amendment to the regulation permitting "Gamma Radiation for the Processing of Food," as published in 33 FR 1465, to include processing by sealed sources of gamma radiation having an energy level of less than 2.2 Mev		Designates AEC letter request as FAP No. 1226
FAP 1233 filed Sept. 13, 1963	Hazleton Laboratories jointly on behalf of the AEC and the Department of the Army	A request was made for use of gamma radiation from sealed sources with maximum energy not to exceed 2.2 Mev, to provide an absorbed dose from 75,000 to 500,000 rads, to be safely used for irradiation of citrus fruits for control of spoilage

show marine products to be acceptable under the conditions studied. Large-scale Army troop feeding tests, which will involve more than 400 individuals, are planned at Fort Lee, Va. Irradiated haddock 15 and 30 days old will be compared with its fresh counterpart. This is the first of a series of large-scale tests, which will include crab, shrimp, clams, and oranges.

Petitions

Three petitions have been approved for irradiated foods (Table III-2): (1) bacon by Co^{60} irradiation with a 4.5- to 5.6-Mrad sterilizing dose; (2) insect disinfestation of wheat and wheat products with 20,000 to 50,000 rads; and (3) amendment to the bacon petition to provide for electron-beam irradiation with 5-Mev, or lower, energy electrons.

A petition has been filed by the U. S. Army for the use of gamma irradiation to inhibit sprout development in white potatoes. Another petition requests a regulation permitting safe use of electron-beam irradiation, with a maximum energy of 10 Mev for radiation sterilization of canned bacon. The AEC has filed a petition for radiation treatment of bacon with sealed gamma sources having an energy of <2.2 Mev (e.g., Cs^{137}). Through the Hazleton Laboratories the AEC and the Army jointly requested the FDA to allow irradiation of citrus fruit, to control spoilage, with sealed gamma sources up to 2.2 Mev, providing an absorbed dose of 75,000 to 500,000 rads.

Future Plans

The results obtained in this program show the continued promise of this developing technology, and we are confident of the future commercialization of radiation processing of foods.

The clearance of radiation-sterilized bacon on Feb. 8, 1963, signaled an increased participation by industry—first, by equipment manufacturers and, second, by food processors. However, no commercialization of food radiation processing has yet occurred, and much development work is still required. The AEC will continue to press toward clearance for human consumption of more kinds of food to provide a broader basis for application of the technique.

Summary

The AEC program on radiation pasteurization of food has resulted in development of processes that extend the refrigerated shelf life of some seafoods as much as fivefold. Work is continuing on these seafoods and on certain fruits that also show promise. Irradiation sources have been designed and, in some cases, constructed. Wholesomeness and acceptability tests are continuing prior to industrial introduction of this new method of food preservation.

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Pasteurized Foods, USAEC Report TID-15144, Continental Can Company, Inc., July 31, 1962.

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Section

IV

Isotopes and Radiation Technology

Process Radiation Development

Use of Gamma Radiation To Produce Wood-Plastic Combinations

By D. S. Ballantine[*]

The results[†] are reported here of two years' research on improvement of the properties of wood by impregnation with a monomer and polymerization of the resin *in situ* by means of gamma irradiation. Wood is a readily available material with a porous structure, and its properties can be varied over a wide range by introduction of polymers. So great is the modification of the properties that an entirely new family of materials is obtained. In principle, the technique used in this work is not new, phenol-formaldehyde and similar resins having been used in the past to decrease wood shrinkage.[1] However, the heat and strong catalysts used to induce polymerization in many cases degrade the wood, improving some properties but deleteriously affecting others. Modification of the technique by using gamma irradiation to bring about polymerization permits more uniform treatment of large sections and shorter curing times than with heat; the small amount of radiation required for polymerization, 0.5 Mrad under the most favorable conditions, does not cause radiation degradation.

The work at West Virginia University has now been brought to the point that larger samples are being prepared for more meaningful testing in specific application. This work is being done in cooperation with personnel at the High Intensity Radiation Development Laboratory at Brookhaven National Laboratory.

A very important part of this project is a continuing interest in the kinetic behavior of other monomers, particularly water-soluble monomers, new additives, and new wood-monomer combinations. The essential objective is to reduce the radiation requirement to a minimum so that any economic potential may be realized.

Properties of Wood-Plastic Combinations

All the properties studied, i.e., dimensional stability and water absorption, hardness, compressive strength, static bending strength, and shear strength, were improved by impregnation with resin. The degree of improvement depended on the wood and resin used, the woods being white pine, sugar maple, white oak, and birch; the resins, polymethyl methacrylate (PMMA), polyvinyl acetate (PVA), and polystyrene (PS). The wood-plastic materials are readily machinable, the grain is accentuated, and the finish is satiny.

Dimensional changes resulting from moisture absorption were decreased about 50% in sugar maple by the introduction of 56% PVA (Fig. IV-1*a*); water absorption decreased when PMMA was introduced (Figs. IV-1*b* and *c*). In more extensive tests, numerous resins showed an antishrinking efficiency varying from −177 to +21% (Table IV-1). Methacrylic acid and acrylic acid were also tested but are not included in Table IV-1 because after irradiation they swelled considerably and warped so badly that measurements were meaningless. This behavior is

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[*] Brookhaven National Laboratory.

[†] This review is based entirely on data of Kent and associates at the Engineering Experiment Station, West Virginia University, contained in USAEC Report ORO-600, Mar. 1, 1963, and in Progress Report to the AEC Division of Isotopes Development, Aug. 22, 1963.

attributed to the polymers' greater affinity for water, and work is in progress on the use of acrylic and methacrylic acids for their bulking action but modified with other monomers to produce a water-resistant copolymer.

The compressive strength of all wood-plastic combinations tested increased with the introduction of increasing amounts of polymer up to about 0.6 lb of polymer per pound of wood (Fig. IV-2). With further increase in the polymer, the compressive strength appeared generally to approach a limiting value, as might be expected.

Hardness (Fig. IV-3), shear strength (Fig. IV-4), and static bending strength (Fig. IV-5) likewise all improved with increasing amounts of polymer introduced. The improvements in shear strength are especially noteworthy be-

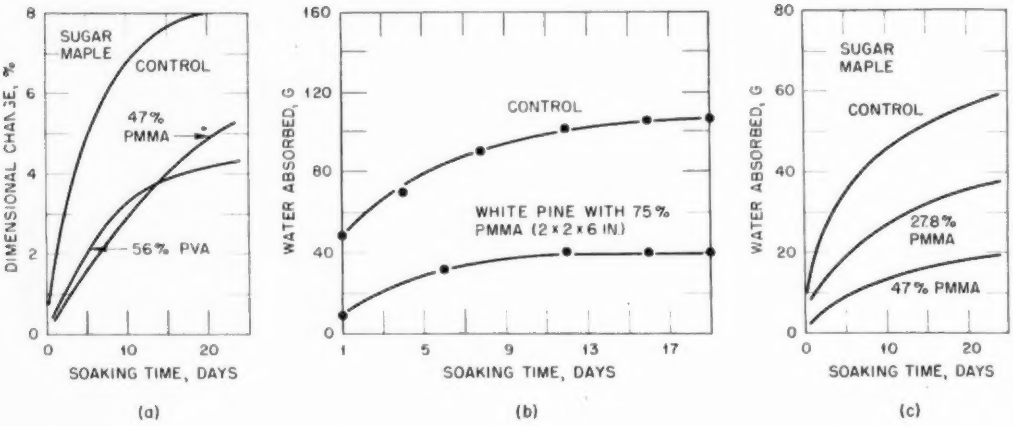

Fig. IV-1 (*a*) Dimensional changes in resin-impregnated wood. (*b* and *c*) Water absorption by resin-impregnated wood.

Table IV-1 DIMENSIONAL STABILITY OF SOME WOOD-PLASTIC COMBINATIONS

		Increase, %						
		Treated samples			Controls			
Monomer	Polymers, wt.%	ΔR	ΔT	ΔV	ΔR	ΔT	ΔV	Antishrinking efficiency, %
Methyl methacrylate	57.7	5.4	8.8	14.7	5.2	8.9	14.6	−0.7
Styrene	57.0	5.2	8.5	14.2	5.1	8.3	13.8	−2.9
Cyclohexyl methacrylate	62.7	5.3	8.7	14.4	5.1	9.2	14.7	+2.0
Amyl methacrylate	61.0	5.2	8.9	14.6	4.8	8.4	13.6	−7.6
Hydroxyethyl methacrylate	71.0	4.6	17.6	23.0	4.6	9.1	14.1	−63.1
Ethylene dimethacrylate	69.0	4.6	7.4	12.4	5.1	8.6	14.1	+12.1
Diaminoethyl methacrylate	64.3	6.3	33.2	41.6	4.8	9.6	15.0	−177.3
Tert.-butylaminoethyl methacrylate	57.8	5.0	13.8	19.5	5.0	9.5	14.9	−30.9
Tetrahydrofurfuryl methacrylate	76.6	4.1	9.1	13.6	4.8	9.0	14.2	+4.2
1,3-Butylene methacrylate	67.2	4.6	9.1	14.1	4.6	9.4	14.5	+2.8
Triethylene glycol dimethacrylate	69.6	4.1	7.3	11.6	4.8	9.5	14.7	+21.1
Tetraethylene glycol dimethacrylate	66.5	4.1	8.2	12.6	4.5	9.1	14.0	+10.0
Butyl acrylate	56.6	3.9	7.2	11.5	4.5	9.7	14.6	+21.2
Ethyl acrylate	56.9	3.6	8.3	12.2	4.3	9.6	14.3	+14.7
Methyl acrylate	66.8	3.2	7.9	11.3	4.4	9.3	14.0	+19.3
2-Ethylhexyl acrylate	65.2	3.8	7.6	11.7	4.6	9.4	14.4	+18.8

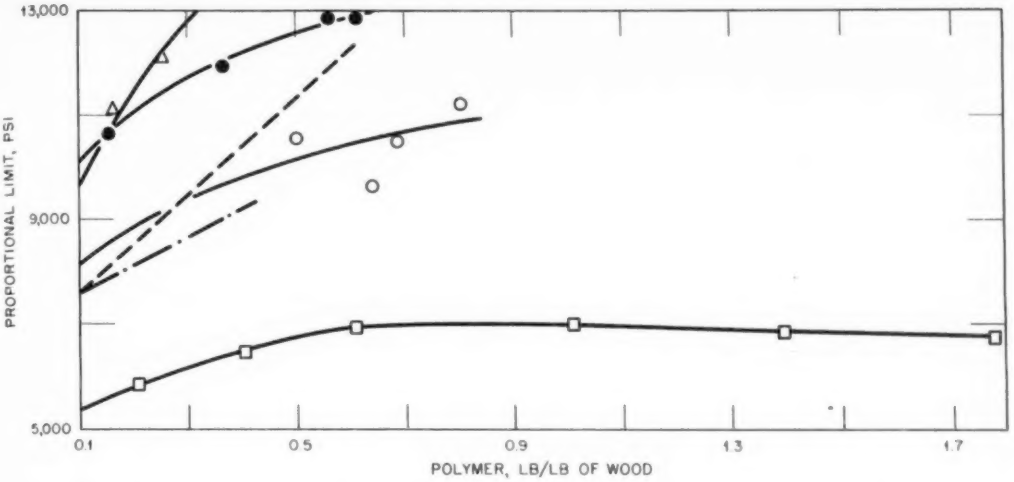

Fig. IV-2 Compressive strengths of wood-plastic combinations. ---, sugar maple with PVA. -·-, sugar maple with PMMA. Δ, white oak with PMMA. ●, birch with PMMA. ○, birch with PVA. □, white pine with PMMA.

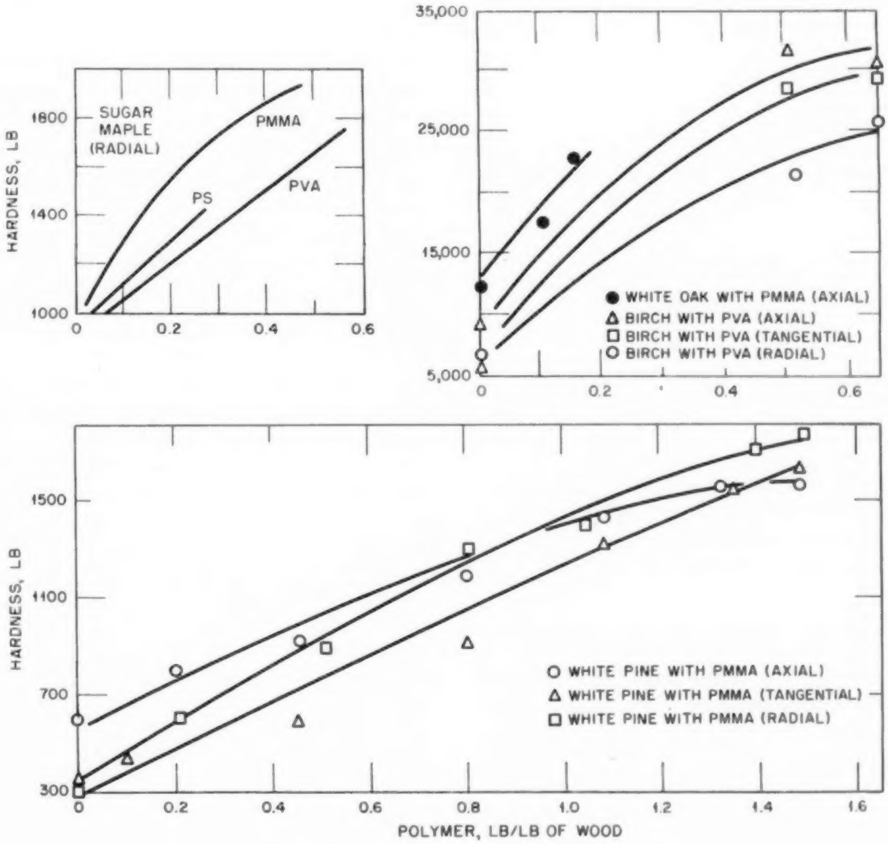

Fig. IV-3 Hardness of various wood-polymer combinations.

attributed to the polymers' greater affinity for water, and work is in progress on the use of acrylic and methacrylic acids for their bulking action but modified with other monomers to produce a water-resistant copolymer.

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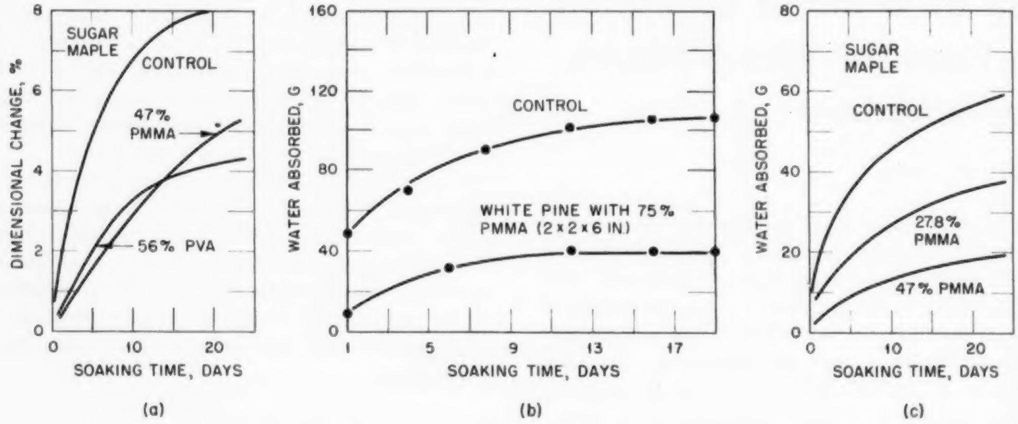

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Amyl methacrylate	61.0	5.2	8.9	14.6	4.8	8.4	13.6	−7.6
Hydroxyethyl methacrylate	71.0	4.6	17.6	23.0	4.6	9.1	14.1	−63.1
Ethylene dimethacrylate	69.0	4.6	7.4	12.4	5.1	8.6	14.1	+12.1
Diaminoethyl methacrylate	64.3	6.3	33.2	41.6	4.8	9.6	15.0	−177.3
Tert.-butylaminoethyl methacrylate	57.8	5.0	13.8	19.5	5.0	9.5	14.9	−30.9
Tetrahydrofurfuryl methacrylate	76.6	4.1	9.1	13.6	4.8	9.0	14.2	+4.2
1,3-Butylene methacrylate	67.2	4.6	9.1	14.1	4.6	9.4	14.5	+2.8
Triethylene glycol dimethacrylate	69.6	4.1	7.3	11.6	4.8	9.5	14.7	+21.1
Tetraethylene glycol dimethacrylate	66.5	4.1	8.2	12.6	4.5	9.1	14.0	+10.6
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Ethyl acrylate	56.9	3.6	8.3	12.2	4.3	9.6	14.3	+14.7
Methyl acrylate	66.8	3.2	7.9	11.3	4.4	9.3	14.0	+19.3
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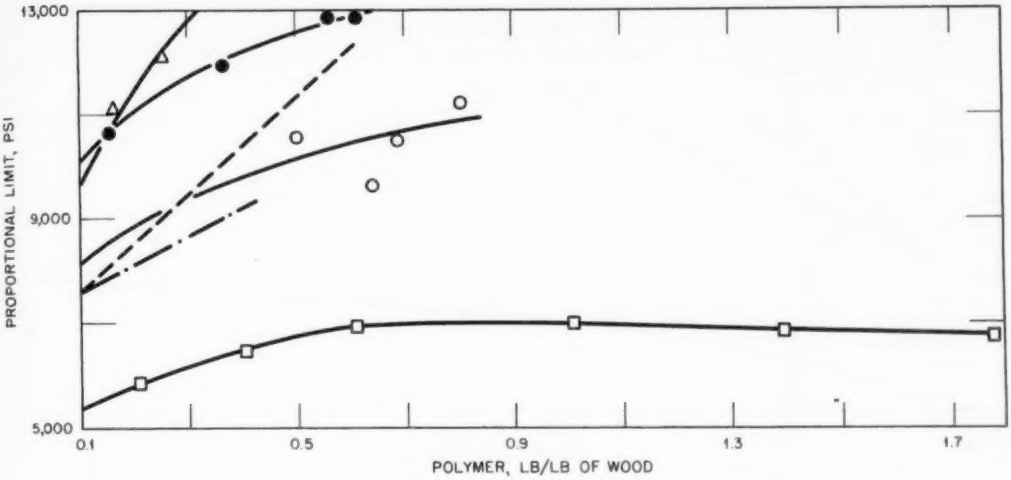

Fig. IV-2 Compressive strengths of wood-plastic combinations. ---, sugar maple with PVA. -•-, sugar maple with PMMA. Δ, white oak with PMMA. ●, birch with PMMA. ○, birch with PVA. □, white pine with PMMA.

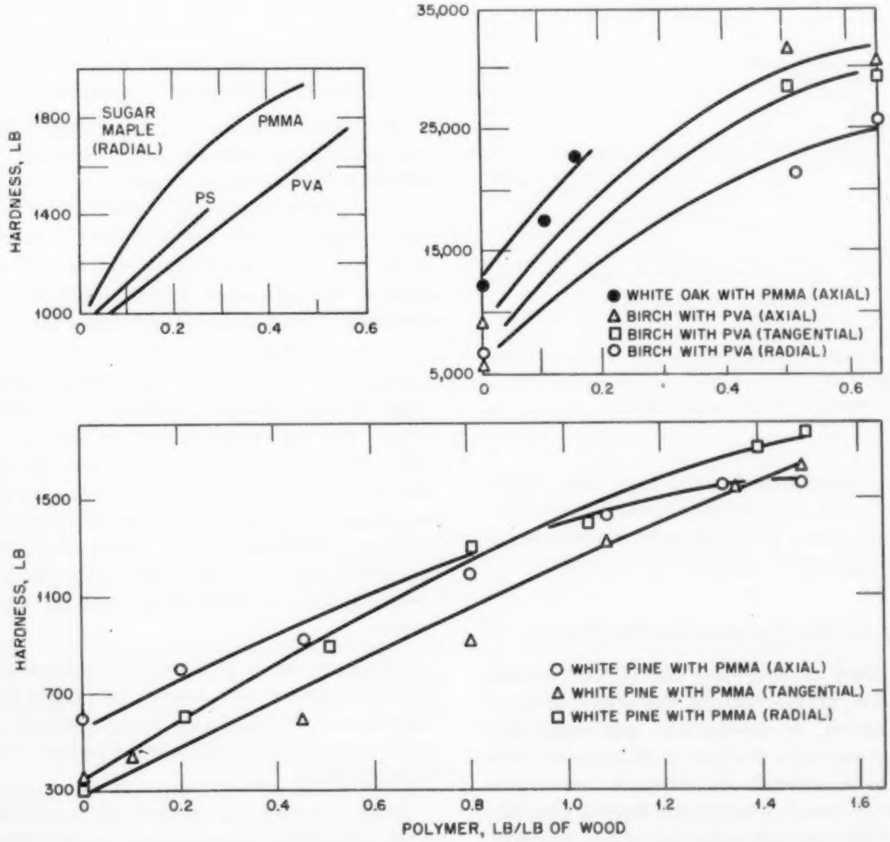

Fig. IV-3 Hardness of various wood-polymer combinations.

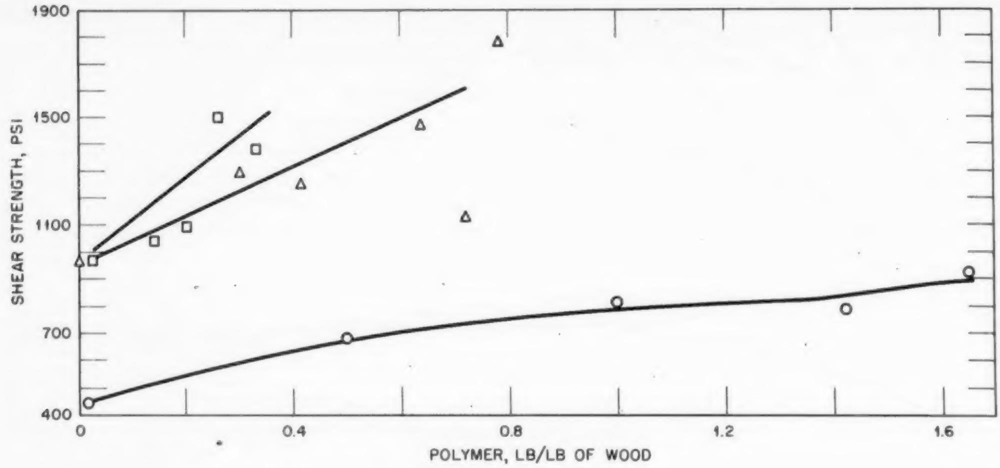

Fig. IV-4 Shear strength, parallel to grain, of various wood-plastic combinations. ○, white pine with PMMA. △, birch with PMMA. □, birch with PVA.

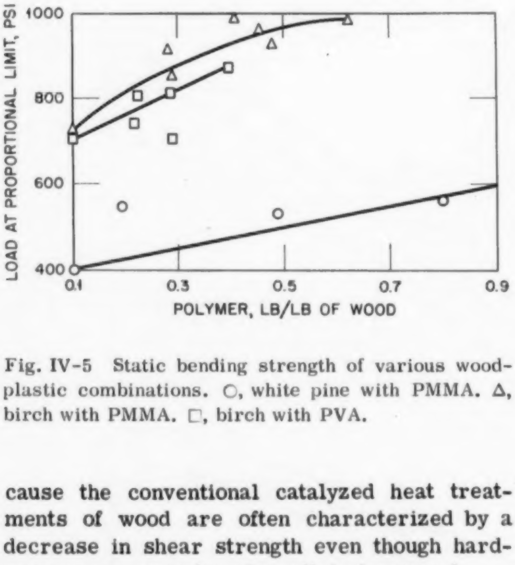

Fig. IV-5 Static bending strength of various wood-plastic combinations. ○, white pine with PMMA. △, birch with PMMA. □, birch with PVA.

cause the conventional catalyzed heat treatments of wood are often characterized by a decrease in shear strength even though hardness or compressive strength is improved.

Chemistry of the Polymerization Process

In an effort to decrease the radiation dose required for polymerization, the effect of various sensitizing, cross-linking, inhibiting, and transfer agents was studied. Although the performance of additives could be quite different in the *in situ* polymerization from that in bulk because of different diffusion rates and solubilities and because of naturally occurring substances in the wood, the results of various experiments showed that in such polymerization the reaction rates and the degree of polymerization can be controlled to a high degree.

Various additives had very small effects on the polymerization of methyl methacrylate in white pine and sugar maple (Fig. IV-6). Reaction in white pine was significantly lower. In these experiments the monomer was not purified, but subsequent experiments (Fig. IV-7) showed the increase in efficiency it might be possible to obtain by the removal of inhibitors.

Curves for polymerization of vinyl acetate in white pine, birch, and sugar maple (Fig. IV-7) also showed slower reaction in the white pine. Also, the inhibiting effect of air is seen in Fig. IV-7*a*, and the importance of monomer purification is readily seen from a comparison of Figs. IV-7*b* and IV-7*d*. Acetic anhydride and maleic anhydride, which were also tested as additives because of their acetylating and/or cross-linking possibilities, had no significant effect.

Little or no grafting of the polymers to the wood occurred, as shown by extraction with appropriate solvents of various woods that had been impregnated with methyl methacrylate and vinyl acetate.

With styrene and methyl methacrylate, carbon tetrachloride produced a significant increase in the polymerization rate and also

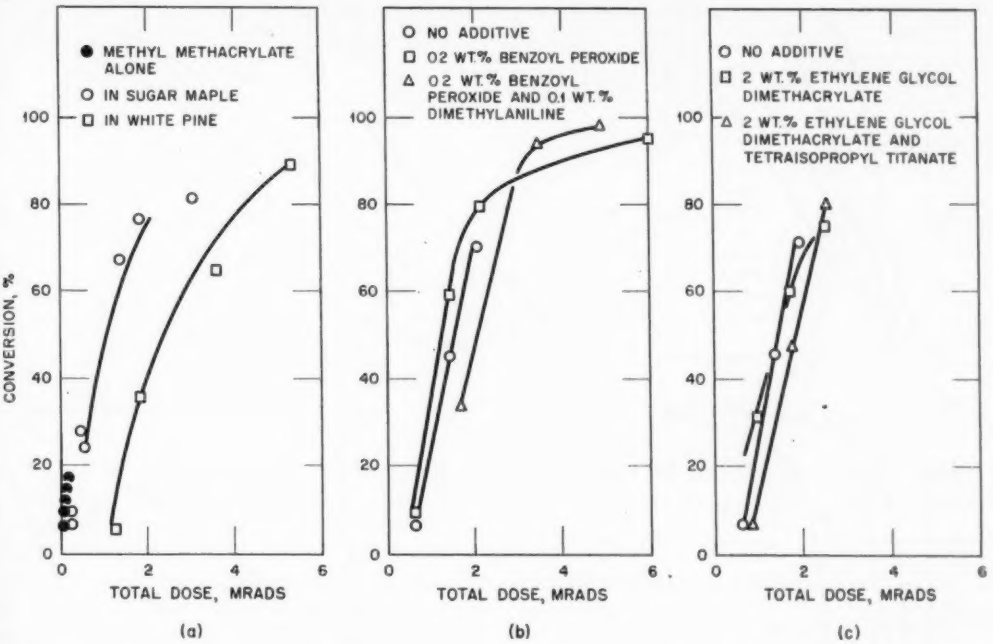

Fig. IV-6 Effect on polymerization of methyl methacrylate of (*a*) total irradiation dose (rate, 20 rads/sec) in sugar maple and white pine and (*b* and *c*) additives in white pine.

Table IV-2 EFFECT OF CARBON TETRACHLORIDE ON POLYMERIZATION OF METHYL METHACRYLATE IN WHITE PINE

Dose rate, rads/sec	Time, hr	Total dose, rads	Conversion,* %			
			MMA		MMA + 10% CCl_4	
30.0	2.0	216,000	0	0	3.8	4.4
	4.0	432,000	0	0	3.0	5.8
	8.2	886,000	9.1	9.2	48.2	56.8
	16.0	1,730,000	19.3	20.9	73.4	75.2
	20.0	2,160,000	67.7	69.0	93.0	100.0
20.8	4.0	300,000	0	0.2	0.5	1.2
	8.25	618,000	0.3	1.3	12.2	13.9
	16.0	1,125,000	1.2	1.2	44.4	51.3
	27.0	2,020,000	48.2	59.7	84.5	90.8
10.0	16.0	575,000	0	0	3.8	4.4
	48.0	1,730,000	20.2	50.0	89.9	84.4

*Determinations in duplicate.

behaved as a chain transfer agent (Tables IV-2 and IV-3). The data in Table IV-3 also show that molecular weights were higher in wood than in the bulk polymerization.

Distribution of Polymer in Wood

Initial studies on methyl methacrylate in white pine indicated that deep penetration of a 2- by 2- by 6-in. block can occur within minutes after contact with a monomer. Some migration of the monomer occurred after impregnation. Although the uniformity of penetration undoubtedly differs greatly with different combinations of wood and plastic, the effect does not at this stage appear to be limiting. The problem of uniformity of penetration, and thus the variation in the final product, is complex

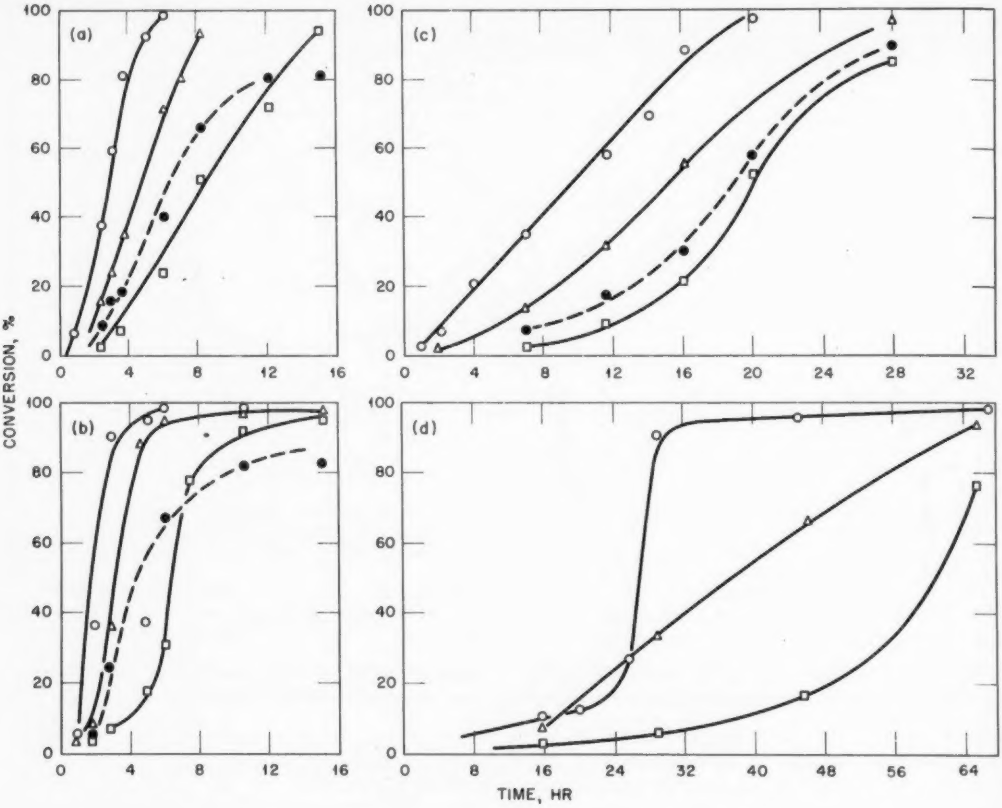

Fig. IV-7 Polymerization of vinyl acetate in (*a*) birch, (*b*) sugar maple, (*c*) white pine (purified), and (*d*) white pine (unpurified). ○, 30 rads/sec, N_2. △, 20.8 rads/sec, N_2. □, 10 rads/sec, N_2. ●, 33.2 rads/sec, air.

because of the effects of size, shape, angle of grain to surface, viscosity of impregnating medium, degree of vacuum applied in evacuation step, soaking pressure, time of soaking, and the nature of the monomer and the wood itself. The problem was studied by a simple method based on density measurements, which was tested and found to be adequate for present purposes.

Experimental Procedure

A Co^{60} gamma radiation source was used. Originally this was a 350-curie unit in the form of a circular array of nine pencils, but later a 10,000-curie source in the form of 40 pencils arranged in two rows was used.

For the initial experiments, samples of selected size were weighed and placed in a vessel to which vacuum was applied and maintained for a time. Monomer was then introduced, and the samples were allowed to soak under atmospheric pressure. An air atmosphere was used initially, but in later work it was replaced with nitrogen. After a predetermined soaking time, the samples were removed, wrapped in aluminum foil to retard evaporation, and weighed. The impregnated wood was irradiated until polymerization was complete. For studying the degree of polymerization vs. dose, the samples were then vacuum dried or placed in a warm air stream until the weight was constant, usually a minimum of 18 hr.

ASTM procedures,[2] modified as necessary, were used to determine the physical properties of the materials produced.

In the more extensive tests on dimensional stability (Table IV-1), samples were prepared from defect-free flat-grain sugar maple sap-

Table IV-3 MOLECULAR WEIGHTS OF POLYMETHYL METHACRYLATE

Type of polymer	Wood	Dose rate, rads/sec	Time, hr	Conversion, %	$[\eta]$*	Molecular weight
Bulk†		30.0	3.5	20.0	0.763	300,000
Bulk†		20.8	4.2	19.0	0.890	370,000
Bulk†		10.0	4.2	7.8	1.122	500,000
Bulk‡		20.8	21.0	32.2	0.826	340,000
Bulk‡,§		20.8	9.0	40.5	0.600	220,000
Wood‡	White pine	30.0	20.0	28.0	1.336	630,000
Wood‡,§	White pine	30.0	20.0	79.0	0.897	380,000
Wood‡	Sugar maple	10.0	65.0	83.8	1.547	770,000
Wood‡,§	White pine	30.0	20.0	93.0	1.052	480,000

* Limiting viscosity, in deciliters per gram.

† Purified monomer, N_2 atmosphere.

‡ Unpurified monomer, air atmosphere.

§ Plus 10% carbon tetrachloride.

wood, $1\frac{1}{4}$ in. in both the radial and tangential directions and $\frac{1}{2}$ in. thick in the direction of the grain. The samples were equilibrated to a moisture content of 7 to 8% in a constant temperature and humidity environment. Three end-matched samples were then treated with each monomer, two others being used as controls. The controls were weighed and measured in the radial and tangential directions with a dial gauge graduated in 0.001 in. One control was retained as the standard, and the other accompanied the three treated samples through the irradiation cycle and testing.

The results (Table IV-1) were calculated as follows:

$$\text{Wt.\% polymer} = 100\ (W_t - W_c)/W_c$$

where W_t is the bone-dry weight of the treated sample combination and W_c is the calculated bone-dry weight of the sample before treatment.

Percent swell in radial direction

$$= \Delta R = 100\ (S_R - D_R)/D_R$$

where S_R is the water-soaked radial dimension and D_R is the bone-dry radial dimension.

Percent swell in tangential direction

$$= \Delta T = 100\ (S_T - D_T)/D_T$$

where S_T is the water-soaked tangential dimension and D_T is the bone-dry tangential dimension.

Percent volumetric swell = ΔV

$$= \Delta R + \Delta T + (\Delta R)\ (\Delta T)/100$$

Percent antishrink efficiency

$$= 100\ (\Delta V_m - \Delta V_u)/\Delta V_u$$

where ΔV_m is the volumetric swell of the wood-plastic combination and ΔV_u is the volumetric swell of control.

Summary

Impregnation of wood with several monomeric resins and polymerization of the resin *in situ* by gamma irradiation resulted in marked improvement of the dimensional stability and water-absorption properties; compressive, static bending, and shear strengths; and hardness.

References

1. *a.* Forest Products Laboratory, Resin Treated Wood, Report No. 1380, 1955.
   b. H. K. Burr and A. J. Stamm, Report No. 1384, Forest Products Laboratory and U. S. Department of Agriculture, 1945.
2. ASTM Standards, Part 6, 1958.

Section V

Isotopes and Radiation Technology

Source Development

Tests on Teletherapy Sources Sealed with Lead O-Rings

By K. W. Haff

Eighteen lead O-ring Co^{60} teletherapy sources from five to eight years old have been tested in the Oak Ridge National Laboratory (ORNL) Source Safety Testing Program to determine the effects of age on them and precautions that would be required in their reuse. The lead O-ring teletherapy source is one of the original concepts of large radioactive source design. Its design evolved from meetings sponsored by the Oak Ridge Institute of Nuclear Studies (ORINS)[1,2] in October and November 1953. The source design adopted by members of the X-ray industry and the Canadian and U. S. Atomic Energy Commissions at ORINS[3] on Nov. 6, 1953, was modified only slightly after that meeting. This source has not been manufactured by ORNL since 1957 because technological advances in producing welded capsules warranted a change to better methods of sealing capsules.

Description of Source

The source capsule consists of five parts: the body, the plug, the cup, and two lead O-rings (Fig. V-1). The body, constructed of an alloy that is 90% tungsten, 6% nickel, and 4% copper, is 1.870 in. high and has an outside diameter of 1.994 in. and an inside diameter of 1.237 in. One end of the body is threaded with $1\frac{5}{8}$-16 American standard thread to receive the plug. This same end of the body is threaded with 1.996-8, 1.9222-1.9031 pitch diameter, general-purpose Acme threads to hold the source in place in a teletherapy machine. The other end of the body has a hole $1\frac{1}{8}$ in. in diameter through which the radiation beam is emitted.

There are three different plugs, also made of the tungsten alloy, for this source. One plug was designed to hold 2-cm-diameter cobalt wafers manufactured in the United States. A second plug held six 1- by 1-cm pellets in the ORINS "cloverleaf" array.[1] The depths of both these plugs could be varied. The third plug, 0.542 in. deep, was used for the Canadian 1- by 1-mm cobalt pellets. The end of the plug is threaded with $1\frac{5}{8}$-16 American standard threads so that it can be screwed into the body. A 45° taper at the end of these threads forms the outer sealing surface. The outer seal ring is a $\frac{1}{8}$-in.-diameter antimony-free lead wire formed into an O-ring having an inside diameter of $1\frac{3}{8}$ in.

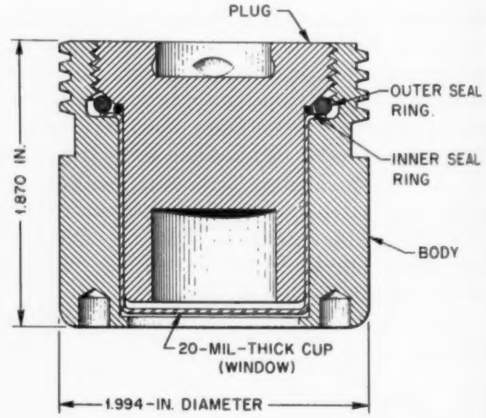

Fig. V-1 Cobalt-60 teletherapy source capsule. Plug and body are made of tungsten alloy (90% tungsten, 6% nickel, and 4% copper). O-ring seal is made of lead, and cup is made of type 305 stainless steel.

The cup is of 0.020-in.-thick type 305 stainless steel with an inner diameter of 1.187 in.

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and an inner height of 1.306 in. The cup has an open top with a 1.5-in.-diameter lip. The cup is seamless and is annealed at 2000°F and air cooled before and after being formed. The lip on the open top of the cup forms the second or inner sealing surface. This seal is a $^1/_{16}$-in.-diameter antimony-free lead O-ring having an inside diameter of $1^3/_{16}$ in.

Test Series

When the sources were received, visual inspection for damage indicated that the capsules were all in perfect condition. They were then subjected to several leak tests to see if they leaked and to evaluate the leak-test methods. These tests included smear tests with absorbent paper to determine the presence of transferable activity, and weighing, immersing in water at 300 psig for 1 hr, and reweighing (a gain in weight would indicate a leak). The sources were then resmeared with an absorbent paper to determine the presence of transferable activity and placed in air at a pressure of 90 psig for 15 min and then in hot water and observed for bubble formation (a stream of bubbles would indicate a leak). The sources were again resmeared with absorbent paper for transferable activity and vacuum leak tested by immersing them in ethylene glycol and decreasing the pressure to 5 in. Hg abs. (Fig. V-2).

The initial smears showed a transferable activity on the surfaces of 13 of the sources of greater than the 1000 dis/min limit established for shipping of sealed sources at ORNL (Table V-1, Smear 1). When the sources were immersed in water at 300 psig, they showed weight gains in all but three cases, indicating inleakage; when the sources were smeared again, all but three showed a surface activity >1000 dis/min. The hot-water-bubble leak test was not effective for determining the leaks, probably because the air had been displaced by water in the preceding test.

Table V-1 COBALT-60 TELETHERAPY SOURCE TESTS

Source No.	Smear 1,* dis/min	Second weight, g	First weight, g	Weight gain, g	Smear 2,* dis/min	Bubbles	Smear 3,* dis/min
1	60	1314.60	1313.85	0.75	10	None	13,570
2	600	1344.50	1345.40	−0.90	2,300	Few	9,890
3	60	1338.45	1337.60	0.85	10	None	42,070
4	1,750	1342.70	1342.65	0.05	406,180	None	727,630
5	6,000	1348.05	1346.49	1.56	12,700	None	109,340
6	27,700	1317.30	1316.40	0.90	147,300	Many	35,550
7	12,674	1314.20	1314.75	−0.55	155,180	Many	102,000
8	250	1300.00	1299.81	0.19	Background	None	105,800
9	2,000	1358.70	1357.85	0.95	19,800	None	18,430
10	8,960	1340.00	1337.85	2.15	270,140	Many	552,140
11	8,310	1354.30	1351.60	2.70	16,000	Few	377,820
12	17,500	1340.05	1338.20	1.85	8,600	Few	142,780
13	143,540	1194.20	1194.10	0.10	77,120	Many	23,810
14	203	1191.10	1188.35	2.75	155,780	Small for 10 sec	124,850
15	5,470	1267.30	1264.48	2.82	7,140	None	141,110
16	39,420	1317.00	1317.90	−0.90	51,090	Many	22,760
17	6,330	1311.85	1311.10	0.75	310,300	None	298,420
18	1,000	1317.60	1315.60	2.00	99,600	Many	178,460

*Smear taken of total source area.

The sources were then leak tested by immersing them in ethylene glycol and decreasing the pressure above the glycol to 5 in. Hg abs. In every case, leaks were present, as indicated by the formation of bubbles.

When the sources were disassembled, each showed breaks in both O-ring seals, with spaces of ~$^1/_{16}$ in. between the ends (Fig. V-3). The condition of the seals was such that the ends appeared to have been pulled apart in the initial sealing process. No other damage was observed. Although all the 18 sources tested were stored in the same container, there was no evidence of

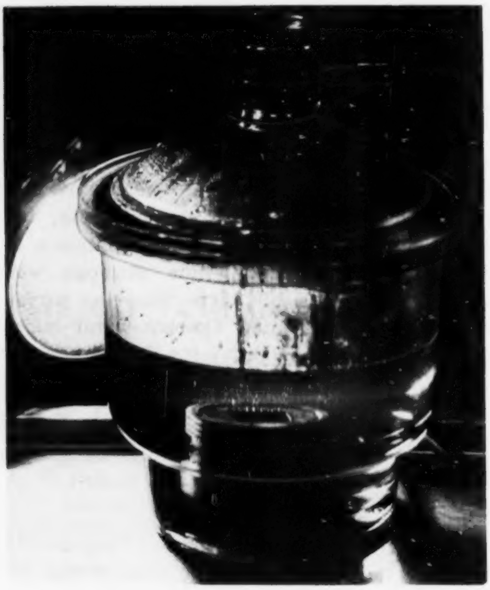

Fig. V-2 Teletherapy source undergoing vacuum leak test.

Fig. V-3 Disassembled teletherapy source showing separation of lead gasket. Ruler is marked in inches.

their gaskets melting. When water was put in the sources, no steam arose, although they dried rapidly. All the capsules, when opened, contained a fine powder that had the appearance of cobalt oxide; however, the powder was not analyzed. This powdery material was removed from five of the sources and weighed. In each case the weight of the material was <1% of the total weight of cobalt in the source. Three of these five sources contained nickel-plated cobalt, and the other two, bare cobalt metal.

Four of the sources were reassembled without cobalt and subjected to structural tests to determine the maximum physical accident a source of this type would withstand. The first unit was dropped onto a 0.25-in.-diameter rod, with the 20-mil-thick window end of the source striking the end of the rod. The first drop was from a height of 6 in., and subsequent drops

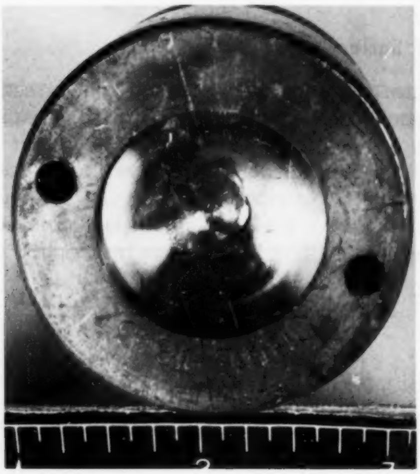

Fig. V-4 Teletherapy source that has been dropped from a height of 2 ft onto a $^1/_4$-in.-diameter rod. Ruler is marked in inches.

Fig. V-5 Teletherapy source cracked by 84,000-lb force. Note crack through "L" in ORNL. Ruler is marked in inches.

were made from 6-in. height increments up to 2 ft, at which drop height the window ruptured. The second and third sources were dropped initially from a height of 2 ft, and in both cases the window was ruptured (Fig. V-4).

The fourth capsule was placed on its side in a 50-ton press, and force was applied until the body cracked. This occurred at a total applied force of 84,000 lb (Fig. V-5).

Summary

The results of tests on 18 five- to eight-year-old teletherapy sources indicate that the sources would withstand considerable physical shock. However, since they all leaked, their lead O-ring gaskets were in poor condition, and transferable contamination was high, it was recommended that sources of this type be re-encapsulated in an all-welded stainless-steel can before being reused.

References

1. Minutes of the Joint Meeting of the Oak Ridge Institute of Nuclear Studies, AEC Isotopes Division, and the X-ray Industry on Teletherapy and Human Radiographic Problems with Isotopes, held Oct. 3, 1953, at Oak Ridge, Tenn.
2. M. Brucer, Chairman, Minutes of the Second Joint Meeting of the X-ray Industry, Oak Ridge Institute of Nuclear Studies, AEC Isotopes Division, Atomic Energy of Canada Limited, and Oak Ridge National Laboratory, held Nov. 6, 1953, at Oak Ridge, Tenn.
3. M. Brucer, A Standard Cobalt-60 Teletherapy Source Capsule, 1953. (Unpublished)

Section

VI

Isotopes and Radiation Technology

Safety

Mexican Co^{60} Radiation Accident

By Gould A. Andrews*

A report[1] from the Mexican National Nuclear Energy Commission, dated November 1962, tells the story of a tragic radiation accident. This report has only recently come to the attention of interested persons in the United States. Shortly after a young Mexican couple and their two children, a 10-year-old boy and a three-year-old girl, moved into a house in March 1962, the boy discovered a 5-curie source of Co^{60} in a field, although no one recognized it as such at that time. It presumably had been in a shielded container, but just how it was removed from the container is not known. The boy carried the source in his pocket for several days, until the first of April, when it was placed in a cabinet that held kitchen utensils near the entrance to the home. The source apparently remained in this cabinet until it was recovered by its owner on July 22, 1962.

Early in April 1962 the boy developed erythema and tissue damage in the abdominal and scrotal regions, with secondary infection. He received medical attention on April 7, and, on April 29, he died as a result of severe damage to the bone marrow, combined with local necrosis and infection at the site of highest radiation dose; however, the fact that these symptoms were due to radiation was not recognized.

The 27-year-old mother, who was pregnant, was exposed to the source while it was in the house, and she became ill somewhat later, around the middle of July, with extensive hemorrhages. She was also found to have severe depression of the blood-cell-forming tissues. She died on July 19, and an autopsy was performed. Again, some of the symptoms of radiation exposure were observed, but radiation exposure as the causative agent was not recognized.

The three-year-old daughter became ill on May 2 with what was thought to be a skin rash, and she later developed a respiratory infection. The child was found to have severe anemia, leukopenia, and thrombocytopenia; she died on August 18. On this day the cause of the deaths was recognized.

The grandmother, who on April 22 had come to visit the family in order to take care of the ailing boy, became ill early in August with what was thought to be hypoplastic anemia. When radiation exposure was identified as the causative mechanism, she was transferred to the National Medical Center, where, in spite of careful studies and vigorous treatment for bone-marrow aplasia, she expired on October 15.

The father had a mild anemia and some darkening of the fingernails, but no more serious

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*Dr. Gould A. Andrews is now Chairman of the Medical Division of the Oak Ridge Institute of Nuclear Studies (ORINS), with which he has been associated in several capacities since 1949. He received the A.B. and M.D. degrees from the University of Michigan and was certified by the American Board of Internal Medicine. Prior to his work at ORINS, Dr. Andrews spent two years as a research assistant at Simpson Memorial Institute and taught at the University of Michigan and the Stritch Medical School. His special interests relate to the effects of radiation on the human body, and he has authored numerous papers in his field. Dr. Andrews is a member of the American College of Physicians, the Society for Experimental Biology and Medicine, the American Thyroid Association, and the American Association for Cancer Research.

difficulty, apparently because he was absent from the household during much of each day while at his job.

Attempts were made to estimate the radiation doses; the total values listed were 1200 r for the father, who recovered, and 4700, 3500, 2870, and 3000 r for the son, mother, daughter, and grandmother, respectively. These dose estimates are uncertain because neither the number of hours of exposure nor the distance from the source is known with any reasonable accuracy, and it seems clear that no valid inference can be drawn about the effects in relation to dose.

From a medical viewpoint, there are several interesting aspects to this accident. The most important of these is associated with the fact that the radiation exposure was considerably more protracted than has been the case in other acute radiation accident situations. When a previously healthy person is exposed briefly to penetrating radiation, experience has shown that within eight weeks the patient will either be dead or will be showing signs of recovery; a protracted marrow hypoplasia compatible with life is usually not seen. However, in this situation, presumably because the radiation was delivered over a relatively long period of time, a more chronic type of injury was produced. This was seen particularly in the grandmother. She was exposed between April 22 and July 22, and yet she survived until October 15, six months after the beginning of the exposure and three months after the end of the exposure.

Another interesting biological aspect of the episode was that nausea, vomiting, and other gastrointestinal symptoms were not noted during or after the exposure. Furthermore, darkening of the fingernails, which is not usually described as a radiation effect, was noted in all the patients.

Little information is given in the report on the circumstances which allowed the Co^{60} source to be left lying in the field, or on the circumstances surrounding its recovery by the owner on July 22, or why it was some 27 days later before the cause of the illness and deaths in the family was discovered. This important part of the episode should serve as a warning to others handling industrial sources. Such accidents seem much less excusable than those which have occurred in reactor operations and other nuclear facilities, where their prevention is technically much more difficult.

Reference

1. First Report on a Radiation Incident, National Nuclear Energy Commission of Mexico, November 1962.

Section

VII

Isotopes and Radiation Technology

Miscellaneous Items of Interest

Radioisotopes Conference

The Second Annual Oak Ridge Radioisotopes Conference will be held in Gatlinburg, Tenn., April 20-22, 1964. (These dates were reported as April 19-21, 1964, in the Fall issue of *Isotopes and Radiation Technology*.) This conference will cover the use of radioisotopes in biology and medicine as well as in industry and research.

New ORINS Radioisotope Training Schedule

The AEC and the Special Training Division of the Oak Ridge Institute of Nuclear Studies announce the 1964 schedule of classes designed to meet the growing demand for personnel skilled in the use of radioactive materials. Up to 48 participants will be accepted in the four-week course in radioisotope research techniques and the three-week engineering courses. Courses of varying duration are offered in specific areas of interest in other fields.

The following is a list of 1964 courses and course descriptions:

ACTIVATION ANALYSIS — This course, designed to provide experience in the application of nuclear techniques in elemental analysis, is available to participants with some background in the handling of radioisotopes. The course emphasizes gamma-ray spectrometry and incorporates experiments using reactors, particle accelerators, and isotopic sources. Both nondestructive and radiochemical techniques are employed. Activation Analysis Course No. 2 will be offered April 6 to 17.

BASIC RESEARCH COURSE — This course trains qualified individuals in fundamental techniques and procedures necessary for the safe use of radioisotopes. Basic Research Course No. 93 will be held January 6 to 31; No. 94, February 10 to March 6; No. 95, April 27 to May 22; and No. 96, September 14 to October 9.

HIGHWAY ENGINEERING COURSE — This course acquaints qualified engineers with theoretical and practical applications of radioisotope techniques for road construction purposes. Among the subjects covered are the uses of radioisotopes in gauging soil density, soil moisture content, and the grade and composition of concrete and other structural components. Highway Engineering Course No. 2 will be held March 9 to 27.

MEDICAL QUALIFICATION COURSE — This course provides basic training and clinical experience necessary for physicians to qualify for a special license issued by the AEC to use radioisotopes on humans. The course consists of three weeks (Basic Medical Week, Preclinical Week, and Clinical Week), offered either in three consecutive weeks or spaced in single weeks for the convenience of the physician.

Basic Medical Week No. 7 will be held January 6 to 10; No. 8, March 2 to 6; No. 9, May 4 to 8; and No. 10, September 14 to 18.

Preclinical Week No. 6 will be held January 13 to 17; No. 7, March 9 to 13; No. 8, May 11 to 15; and No. 9, September 21 to 25.

Clinical Week No. 4 will be held January 20 to 24; No. 5, March 16 to 20; No. 6, May 18 to 22; and No. 7, September 28 to October 2.

NUCLEAR GEOLOGY COURSE — This course provides the researcher with information and practice in the current techniques applicable to geological and geochemical research. The course is tentatively scheduled for fall 1964.

Stable Isotopes

The following were added to the ORNL inventory of stable isotopes available for sale:

~20 g	>95% Cd^{113}
~1 g	79% Cd^{114}
~7 g	95% Sm^{150}
~0.3 g	45% Te^{120}

In December 1963 the concentration of Ne^{21} in the separation process was 32% and Ar^{31} was 22%. These isotopes are available in very limited amounts.

AEC Changes Co^{60} Price Schedule

The AEC has changed its schedule of prices for large-quantity purchases of Co^{60}. The change from schedules announced in May 1961 is made to encourage bulk purchase of Co^{60} and to stimulate wider use of this radioisotope for industrial processes.

The price schedules, effective Sept. 10, 1963, and applicable discounts are given below.

Specific activity, curies/g	List price, per curie	
	Old	New
1 to 25	\$2.00	Same
26 to 40	3.00	Same
41 to 55	4.00	Same
56 to 70	6.00	\$5.25
71 to 85		5.65
86 to 100		5.85
101 to 115		6.05
116 to 130		6.20
131 to 145		6.40
146 to 160		6.60
161 to 175		6.80
176 to 190		7.00
191 to 205		7.20

Discounts

Quantity, curies	Old	New
0 to 5,000	List price	Same
5001 to 25,000	List price less 15%	Same
25,001 to 100,000*	List price less 30%	Same

*Cobalt-60 with a specific activity of 30 curies/g or less will be priced at \$0.50 per curie for custom orders >100,000 curies.

High-Radiochemical-Purity I^{131} and Xe^{133}

Iodine-131 with a radiochemical purity of 99.9999% and Xe^{133} with a radiochemical purity of 99.998% (exclusive of Xe^{131} and Kr^{85}) were obtained at Oak Ridge National Laboratory (ORNL). A 2.5-g 93% U^{235}-aluminum Oak Ridge Research Reactor fuel ring was heated at 670°C under 100-μ Hg pressure, and the iodine and xenon thus released were separated in a series of temperature-controlled cold traps.

Shutdown of Graphite Reactor Necessitates Shift in Reactor Services

The ORNL Graphite Reactor, shut down on Nov. 4, 1963, was used in the first commercial production of radioisotopes and has been the workhorse of the isotope production and reactor services program for many years. With this shutdown ORNL will offer activations and service irradiations only in the Low-Intensity Test Reactor [flux $\sim 5 \times 10^{13}$ neutrons/(cm^2)(sec)] and the Oak Ridge Research Reactor [flux $\sim 1 \times 10^{14}$ neutrons/(cm^2)(sec)].

AEC Withdraws from Routine Production of Radioiodine

The AEC withdrew from routine production and distribution of I^{131} and I^{125} on Oct. 1, 1963. This is in accordance with the Commission's general policy to discontinue providing material or services that are reasonably available from commercial sources. The Commission's decision was based on consideration of these particular isotopes only.

The Commission will continue to supply its high-specific-activity I^{131} to meet special research and other needs until an equivalent product is available commercially. The price of this I^{131} will be continuously adjusted to recover full production cost or to the lowest cost of commercially supplied I^{131}, whichever is higher. At present, existing price schedules will be continued. The AEC, as a part of its broader isotopes development program, will continue research leading toward a large-scale I^{125} production technology, which would then be made available to industry.

Commercial suppliers of radioiodine include Abbott Laboratories, Oak Ridge, Tenn.; General Electric Company, Pleasanton, Calif.; Iso/Serv, Inc., Cambridge, Mass.; Nuclear Consultants

Corporation, St. Louis, Mo.; Nuclear Science and Engineering Corporation, Pittsburgh, Pa.; E. R. Squibb and Sons, New Brunswick, N. J.; Union Carbide Corporation, Tuxedo, N. Y.; and Volk Radiochemical Company, Skokie, Ill.

Remote Arctic Weather Station Still Operating

A 5-watt $SrTiO_3$-fueled thermoelectric generator, described in *Isotopes and Radiation Technology*, 1(1): 89 (Fall 1963), is still successfully operating an automatic meteorological data-transmitting radio station on Axel Heiberg Island in the Arctic. The 17,500-curie source and electronic system, designed, manufactured, and tested by the Martin Nuclear Division and fueled by ORNL, has already outlived its minimum life expectancy of two years and continues to telemeter back to earth, at 3-hr intervals, wind direction and speed, barometric pressure, and temperature.

AEC Simplifies Regulations on Licensing of Radioisotopes as Tracers

The AEC has simplified its regulations on licenses required to introduce small quantities of radioisotopes into products to serve as tracers. In the past, Commission regulations did not expressly state whether a company must be licensed before it could engage another firm to perform radiotracer services for it, even though the radiotracer firm is licensed for the purpose. Such dual licensing has been found to be unnecessary from the standpoint of maintaining radiation safety. Licenses issued to firms that perform radiotracer work prescribe conditions that must be met to ensure safety.

Under the amendment to AEC regulations, firms that provide radiotracer services resulting in license-exempt concentrations of radioisotopes in products will continue to be licensed, but firms for which the service is performed will not be required to have a Commission license. Annual reports will be required from radiotracer firms, and firms wishing to perform radiotracer services on their own products must be licensed by the Commission. There is no requirement that firms use the services of another company or person for radiotracer work, provided they have a license from the AEC to perform this work themselves.

The amended regulation also contains provisions under which products containing radiotracers added under licenses of states that have agreements with the Commission may be distributed with exemption from Commission licensing procedures. The exemption would not authorize transfer of radiotracers in any food, beverage, drug, or other product designed for ingestion or inhalation by, or application to, a person.

New Edition of Heath's Spectrum Catalog Available

The new edition of the *Scintillation Spectrometry Gamma-Ray Spectrum Catalogue* by R. L. Heath, Phillips Petroleum Company, Atomic Energy Division, Idaho Falls, Idaho, went to the printer in October 1963 and was to become available by the middle of January 1964. The new edition of the catalog, which has the number IDO-16880 and is in two volumes, will receive standard AEC distribution under the category of Physics and Mathematics and will be available to others from the Office of Technical Services, U. S. Department of Commerce, Washington, D. C., 20230. The new edition of the report will be in loose-leaf form, and supplementary sheets will be available. The method for distribution of these supplementary sheets has not yet been determined.

U. S. Papers for Third United Nations Geneva Conference

Several papers of interest to workers in the isotope field were chosen by the AEC for presentation by the United States at the third Geneva Conference in September 1964. These are as follows:

General Session F. Applications of Isotopes and Radiation Sources in the Physical Sciences

Advances in Utilization of Isotopes in Science and Industry	P. C. Aebersold, AEC
Advances in Neutron Activation Analysis	V. Guinn, General Atomic
Large Sources for Chemical Irradiation Processes	J. E. Machurek, AEC
Use of Tracers in Hydrology	A. Clebsch, U. S. Geological Survey

General Session . Applications of Isotopes and Radiation Sources in the Life Sciences. Radiobiology

Tracers and Radiation in Agriculture	B. Schweigert, Michigan State University

Session 1.6. Direct Conversion of Heat to Electricity

Isotopic Power Sources Coupled with Thermoelectric Converters	J. Morse, Martin Co.
Reactor Direct Conversion Units Including SNAP	H. Dieckamp, Atomics International
Thermionic Nuclear Reactors	G. Grover, Los Alamos Scientific Laboratory

Session 2.7. Fuel Reprocessing

Large-Scale Fission-Product Recovery	R. Tomlinson, Hanford Atomic Products Operation

Session 3.6. Nuclear Safety Research

Aerospace Safety of Isotopic and Reactor Power Sources	G. Fowler, Sandia

Session 3.9. Isotope Separation

Heavy-Water Separation	W. P. Bebbington, Du Pont, Savannah River Laboratory

Session 3.10. Miscellaneous

Project Plowshare Experiments	G. Johnson, Livermore
Chemonuclear Process and Technology	B. Manowitz, Brookhaven National Laboratory

Additional Radioactive Cobalt To Be Produced for Sale

Approximately 465,000 curies of Co^{60} approved for production at the AEC's Savannah River Plant, Aiken, S. C., will be distributed by ORNL. The specific activity of the radioactive cobalt will range between 400 and 500 curies/g, the highest ever produced. Delivery is scheduled for 1966. The Commission's Division of Production in Washington authorized the irradiation of the cobalt in a Savannah River reactor upon request of the Division of Isotopes Development.

The material will be made available through ORNL to a number of organizations for experimentation in radiography, for teletherapy, as the heat source in a thruster in space vehicle application, and for electric generators. The high specific activity of the material will also be evaluated in the general area of radiation processing.

AEC's High Intensity Radiation Development Laboratory Dedicated

The High Intensity Radiation Development Laboratory, one of the world's most versatile gamma-radiation facilities, was dedicated Nov. 22, 1963, at Brookhaven National Laboratory, Upton, Long Island, N. Y. This facility is designed to advance the program for process radiation development by: (1) obtaining engineering data on a variety of radiation sources in the megacurie range; (2) developing more efficient techniques for handling large-scale radiation sources; and (3) training scientists and engineers in the uses of such radiation sources for research purposes and ultimately for industrial applications. This information is essential for the design of future irradiation facilities. Those currently under development are for the production of plastics and chemicals, cold sterilization of drugs and medical supplies, and pasteurization of perishable foods.

The cost of the facility, including equipment, was $1,880,000. The total area of the building is approximately 15,000 sq ft. Ground was broken for the laboratory on Nov. 28, 1960.

Dr. Gerald F. Tape, member of the AEC, spoke to those attending the ceremony. Dr. Tape was a senior member of the Brookhaven staff from 1951 to 1962. As Deputy Director and then as Acting Director of the Laboratory, he took an active interest in the development of this important new research facility. Dr. Paul C. Aebersold, Director of the Commission's Division of Isotopes Development which administers the program, participated in the ceremonies.

Savannah River Plant To Produce Curium-244

The Savannah River Plant will begin to produce Cm^{244}, a radioactive material potentially useful as a long-lived thermal-energy source in space devices. The 1964–1966 program will produce around 3000 g of Cm^{244}. Curium-244 will be produced in the Savannah River reactors by extended irradiation of plutonium, during which plutonium isotopes are burned out and subsequently converted to the element americium, which in turn is converted to Cm^{244}.

The plant also has produced another radioactive material, Pu^{238}, for applications in space devices. Plutonium-238 is being used to supply the energy that is converted into electricity to generate signals from two satellites now in orbit.

Two Nuclear-Powered Satellites Still Operating Successfully

The nation's first two satellites wholly powered by nuclear energy were launched recently from Vandenberg Air Force Base, Calif. Signals from the satellites, boosted into orbit by Air Force Thor Able Stars, are being transmitted successfully with electricity from isotopic power generators developed by the Nuclear Division of the Martin Co. for the AEC. These nuclear power devices, designated SNAP-9A, are the first use of all-nuclear power systems on satellites.

SNAP-9A is designed to provide 25 watts of direct electric current from Pu^{238}. Two similar but smaller generators, the world's first and second devices using nuclear power in space, were placed in orbit in Department of Defense satellites in 1961. Those satellites derived part of their electric power from solar cells.

ORNL Isotope Shipments Decrease

Reflecting AEC's deemphasis of Commission sponsorship of production programs and its increased emphasis on research and development activities to improve production techniques, isotope shipments from ORNL decreased from 12,864 shipments in 1962 to 12,520 in 1963.

High-Enrichment Isotopes Inventoried

In January the Ne^{21} content of the enriched material being produced by thermal diffusion at ORNL reached 31%. ORNL's electromagnetic separations facility for stable isotopes has recently inventoried gram quantities of 94% Te^{122}, 93.9% Te^{124}, 95% Te^{125}, 97% Te^{126}, 99.1% Ti^{48}, and 97.2% Cd^{116}.

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